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<td>Andrea Juliana Hernandez Villamizar (Early Career Scientist), Universidad Nacional de Colombia, Department of Chemical and Environmental Engineering, Bogota D.C., Colombia</td>
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<td>Scott Eilerman (Early Career Scientist), NOAA ESRL Chemical Sciences Division / Cooperative Institute for Research in Environmental Sciences (CIRES), Boulder, CO, USA</td>
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<td>Luke Schiferl (Early Career Scientist), MIT, Dept of Civil and Environmental Engineering, Cambridge, MA, USA</td>
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<td>PRAPHULLA CHANDRA BOGGARAPU (Early Career Scientist), Department of Earth and Environmental Sciences, Indian Institute of Science Education and Research Mohali, Sector 81, S.A. S. Nagar, Manauli PO, Punjab, 140306, India</td>
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<td>David Miller (Early Career Scientist), Institute at Brown for Environment and Society, Brown University, Providence, RI, USA</td>
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<td>Kerneels Jaars (Early Career Scientist), North-West University, Unit for Environmental Sciences and Management, Potchefstroom, South Africa</td>
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<td>Jeffrey A. Geddes (Early Career Scientist), Boston University, Department of Earth and Environment, Boston, MA, USA</td>
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<td>Catherine Scott (Early Career Scientist), Institute for Climate and Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds, UK</td>
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<td>Gilmarie Santos-Figueroa (Early Career Scientist), Department of Chemistry, University of Puerto Rico, San Juan, Puerto Rico</td>
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<td>Distribution of oceanic dimethyl sulfide in the Arctic Ocean and the Southern Ocean made by membrane inlet mass spectrometry</td>
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<td>Rebecca Hornbrook, Atmospheric Chemistry Observations &amp; Modeling Laboratory, NCAR, Boulder, Colorado, USA</td>
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<td>Claudia Steadman (Early Career Scientist), University of Edinburgh and Centre for Ecology and Hydrology, Edinburgh, United Kingdom</td>
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<td>Anke Roiger, Institute of Atmospheric Physics, German Aerospace Centre (DLR), 82234 Oberpfaffenhofen, Germany</td>
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<td>Mauricio Osses Alvarado, Departamento de Ingeniería Mecánica - Universidad Técnica Federico Santa María, Santiago, Chile</td>
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<td>Anna Robertson (Early Career Scientist), University of Wyoming, Atmospheric Science, Laramie, WY, USA</td>
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<td>Sebastian Tolvett (Early Career Scientist), Departamento de Ingeniería Mecánica, Universidad Tecnológica Metropolitana, Campus Macul, Av. José Pedro Alessandri 1242, Ñuñoa, Santiago</td>
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<td>Ravan Ahmadov, Earth System Research Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO, USA</td>
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<td>Peter Brauer (Early Career Scientist), Wolfson Atmospheric Chemistry Laboratories, Department of Chemistry, University of York, York, UK</td>
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<td>yanli ge (Early Career Scientist), State Key Joint Laboratory of Environment Simulation and Pollution Control, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences &amp; College of Resources and Environment, University of Chinese Academy of Sciences, Beijing, China</td>
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<td>yan zhao (Early Career Scientist), State Key Joint Laboratory of Environment Simulation and Pollution Control, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences &amp; College of Resources and Environment, University of Chinese Academy of Sciences, Beijing, China</td>
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Michael Link (Early Career Scientist), Colorado State University

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Renee McVay (Early Career Scientist), California Institute of Technology, Chemical Engineering, Pasadena, CA, USA

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Timothy John Wallington, Research and Advanced Engineering, Ford Motor Company, Dearborn MI 48121, USA

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**Session 6: Atmospheric Chemistry - Observing Composition and Variability**

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On behalf of the Committee on the Future of Atmospheric Chemistry Research

Abstract:

It has been more than thirty years since the field of atmospheric chemistry has evaluated its research accomplishments and future directions. Recognizing that there have been tremendous changes in our understanding of the chemistry of the atmosphere and our ability to investigate it through field, theoretical, laboratory, and modeling efforts over the past three decades, in 2015 the National Science Foundation once again requested that the National Academies undertake a study to identify priorities and strategic steps forward for atmospheric chemistry research for the next decade. The Committee on the Future of Atmospheric Chemistry Research was formed with the task of summarizing the rationale and need for supporting a comprehensive U.S. research program in atmospheric chemistry; commenting on the broad trends in laboratory, field, satellite, and modeling studies of atmospheric chemistry; determining the priority areas of research for advancing the basic science of atmospheric chemistry; and identifying the highest priority needs for improvements in the research infrastructure needed to address those priority research topics.

A central part of the Committee’s activities was to seek the thoughts and advice of the U.S. atmospheric chemistry community on future priority areas in atmospheric chemistry research during a series of “town hall” meetings during the spring and summer of 2015 and through an online portal. Approximately 300 individuals participated in these efforts, providing valuable insights and suggestions to the Committee. After a number of meetings and extensive deliberations, the report The Future of Atmospheric Chemistry Research: Remembering Yesterday, Understanding Today, Anticipating Tomorrow was released in late summer of 2016. The conclusions and recommendations of this report will be presented.
K.002 Aerosols in atmospheric chemistry and biogeochemical cycles.

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Abstract:

Atmospheric aerosols have complex and variable chemical composition and properties depending on their size, their sources and atmospheric ageing. They affect human health, the climate since they interact with radiation and atmospheric water, and the ecosystems because they can carry nutrients or toxic components. Aerosols and associated water also serve as media for multiphase chemistry that changes both atmospheric oxidant levels and aerosol properties. 
Humans have significantly modified the composition of atmospheric aerosol. Desert dust, the most abundant aerosol originating from natural continental sources, interacts with acidic compounds of anthropogenic origin that increase dust solubility and mobilize nutrients carried by dust, such as Fe and P. Organic ligands also interact with dust elements since water soluble organic di-acids present in aerosol water and in cloud droplets can form Fe-complexes, changing dust solubility as well as organics’ chemical properties. Furthermore, the organic pool in the atmosphere has variable content in C, H, O, N and other elements depending on the origin and the history of organics in the atmosphere. Organics are carriers of nutrients and thus key players in the biosphere-atmosphere-climate interactions, while they have a significant anthropogenic component due to primary emissions or interactions with pollution. Remarkably, organic aerosol is found to increase in the atmosphere, contrary to sulfate aerosol that has been mitigated the past decades.

In line with this vision for the role of aerosols in the atmosphere, we summarize recent global chemistry-transport modeling studies focusing on organic and dust aerosol components, which account for multiphase chemistry, aerosol ageing in the atmosphere, nutrients emissions, atmospheric transport and deposition. The importance of human-driven emissions is derived by sensitivity simulations using past and future anthropogenic emissions of pollutants. Uncertainties and implications of our findings are discussed.
Abstract:

Earth’s growing population and increasing climate forcing are leading us to a new regime of impacts that pose unique challenges to society. A key challenge is understanding our environment (namely, the atmosphere and ecosystems) sufficiently well that we can describe the processes that define its present state and project how it might change in the future. Many attending this IGAC meeting are motivated by this need for understanding and have devoted themselves in significant ways. In this new regime, society is faced with new and complex decisions regarding our relation to our environment. As atmospheric and environmental scientists, we increase our effectiveness by being aware of how our environmental research connects to decisions. The best decisions require the fundamental understanding that we are continually seeking, and require us to frame and describe it in a manner relevant for the problems at hand and, above all, to communicate it effectively to stakeholders and decision makers. In this keynote, I will present a few examples from my experience that emphasize this perspective as a way of increasing awareness and providing encouragement for the challenges ahead.
1.004 Air pollution in Rwanda, a growing East African country.

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Abstract:

The Rwanda Climate Observatory Project, located on the summit of Mt. Mugogo, a 2.5 km peak in the Virunga mountain range, has begun measurements of black carbon aerosol concentration, ozone and carbon monoxide gas concentration, and solar intensity. Black carbon levels close to those in major US cities were found in this rural region, likely from local and transported biomass burning. Major sources of air pollution include agricultural burning, cook fires, charcoal making, kerosene lightning, brick kilns, and older diesel generators/vehicles. CO and O₃ measurements were used in conjunction with BC aerosol data and HYSPLIT back trajectories were also used to help discriminate between periods of heavy burning and periods of pollution transport from Kigali, Kampala, and other large East African cities, which may have more black carbon contribution from diesel vehicles and generators. Additionally, low-cost air quality monitors will be deployed in different areas of Kigali, the capitol of Rwanda, to determine approximate PM2.5 concentrations near different sources within the city to compare with rural data. Rwanda is one of the least-urbanized nations in Africa, though the government is pushing for higher urbanization rates. It is also the most densely populated country in Africa, and its rural areas are filled with small households. Currently, initiatives to supply more-efficient cookstoves to lower income households, development of cleaner-burning fuel from recycled agricultural waste, and new regulations on vehicle emissions and importation are underway. These new initiatives seek to help Rwanda grow and urbanize in the greenest way possible; however, little ambient data on local air quality is available in different regions of Rwanda for a baseline study before and after these initiatives. With this work, we seek to understand major pollution sources in the region to increase the efficiency of government policy initiatives.
Modelling urban δ¹³C variations in the Greater Toronto Area.

Early Career Scientist

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Abstract:

Even in urbanized regions, carbon dioxide (CO₂) emissions are derived from a variety of biogenic and anthropogenic sources and are influenced by atmospheric transport across borders. As policies are introduced to reduce the emissions of CO₂, there is a need for independent verification of emissions reporting. In this work, we use carbon isotope (¹³CO₂ and ¹²CO₂) simulations in combination with atmospheric measurements to distinguish between CO₂ sources in the Greater Toronto Area (GTA), Canada. This is being done by developing an urban δ¹³C framework based on existing CO₂ emissions data and forward modelling. We first developed a CO₂ inventory for the GTA at a very fine spatial and temporal resolution (0.02° x 0.02° and hourly, respectively). We compared the outputs from the Lagrangian dispersion model FLEXPART (at 0.1° x 0.1°) and the chemistry transport model GEM-MACH (at 0.02° x 0.02°) to evaluate the impact of model resolution on the produced simulations. These model results are used in our framework in combination with region-specific δ¹³C signatures of the dominant CO₂ sources; the product is compared against highly accurate ¹³CO₂ and ¹²CO₂ ambient data. Locally, anthropogenic CO₂ in urban areas is often derived from natural gas combustion (for heating) and gasoline/diesel combustion (for transportation); the isotopic signatures of these processes were measured to be significantly different (approximately δ¹³CVPDB = -44 ‰ and -29 ‰ respectively in the GTA and can be used to infer their relative contributions. Utilizing our δ¹³C framework and differences in sectoral isotopic signatures, we quantify the relative contribution of CO₂ sources on the overall measured concentration and assess the ability of this framework as a tool for tracing the evolution of sector-specific emissions.
Evaluation of surface emissions in Asia.

Early Career Scientist

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Abstract:

During the past few years, severe air pollution episodes were recorded in China, India and other regions in Asia. In order to understand the origin and evolution of these events, and to perform forecasts of air quality in Asia, it is necessary to have an accurate knowledge of the surface emissions involved in air pollution in this region. During the past few years, several inventories providing anthropogenic emissions for different regions in Asia were developed, as well as global emissions inventories. Depending on the inventory, the emissions are provided for different years during the 1960-2012 period. We will present a comparison of the inventories available for this period for China, India, South-East Asia, Korea and Japan. The comparisons will focus on total anthropogenic emissions and emissions from traffic. Emissions for speciated volatile organic compounds, i.e. alkanes, alkenes, aldehydes and aromatics will also be discussed. Emissions estimates of NO\textsubscript{X}, SO\textsubscript{2} and VOCs for several regions in Asia were also quantified using inverse modeling techniques and satellite observations from the OMI and GOME-2 instruments. These optimized emissions will also be included in the analysis.
Impacts of meteorological factor and aerosol chemical compositions on visibility impairment in Nanjing, China.

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Abstract:

With rapid industrialization and urbanization, air pollution and corresponding visibility problems in Nanjing were increasingly severe during the last few years. To better understand the potential causes of impaired visibility, particulate matter mass and chemical composition were measured from May 2013 to May 2014 in Nanjing. During the period, major water soluble ions and organic carbon exhibited obvious seasonal variations with the highest level in winter. The daily average visibility in Nanjing over study period ranged from 1.2 km to 18.4 km. High aerosol concentration and relative humidity were two important factors that cause low visibility events in Nanjing. The correlation between PM$_{2.5}$ concentration and visibility under different relative humidity values showed that visibility was exponentially decreased with the increased PM$_{2.5}$ concentrations when relative humidity less than 80%. However, the relationship was no longer to follow the exponentially decreasing trend when relative humidity > 80%, and the visibility maintained in very low values, even with low PM$_{2.5}$ concentrations. This indicated the hygroscopic growth of particles played more important roles for reduction of visibility. The annual average chemical extinction coefficient based on the revised IMPROVE equation was 267.69 ± 139.24 Mm$^{-1}$ in Nanjing. On average, organic matter was found to be the largest contributor accounting for 35.69% of chemical extinction coefficient. The contributions of ammonium sulfate and ammonium nitrate were also important with the annual average of 28.80% and 24.08%, respectively. For visibility > 10 km, organic matter was the largest contributor to extinction coefficient, while organic matter and ammonium sulfate were the main contributors for visibility < 5 km. The reduction of carbonaceous species and sulfate could effectively improve the visibility of Nanjing. The paper aims to help environmental scientists and policy makers understand air pollution in Nanjing and air pollution control strategies taken by government.
Chemical Composition of PM10 a background site in Leicester, UK: Correlation between Levoglucosan and PNSD.

Early Career Scientist

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Abstract:

Exposure to particulate matter has been shown to have detrimental effects on health, in particular in vulnerable groups such as the elderly, children, and those with pulmonary or cardiovascular disease.

Particle number size distribution (PNSD) was measured by an ultrafine particle monitor (UFP TSI model 3031), black carbon (BC) was measured by a MAAP (Thermo-5012). Particulate matter samples (PM$_{10}$) were collected from April 2013 to May 2014 at an urban background site in Leicester, UK. In order to characterize the chemical composition, several chemical components were analysed: water-soluble ions (chloride, nitrate, sulphate, sodium, potassium, ammonium, calcium, and magnesium), monosaccharide anhydrides (levoglucosan (Lev), mannosan (Man), and galactosan (Gal)), and the elemental and organic carbon (EC/OC). The measurements were taken at the Automatic Urban and Rural Network (AURN) monitoring site at University of Leicester. The monitoring was performed as part of the EU project Joint Air Quality Initiative (JOAQUIN, www.joaquin.eu) supported by the INTERREG IVB NWE programme. The samples were collected daily (24 hour exposure) onto 47 mm quartz filters (Pall TissuquartzTM filters, 2500 AQT-UP) using a sequential sampler (Leckel SEQ47/50) with PM$_{10}$ inlet, running at 2.3 m$^3$/h for 24 h per filter. Filters were weighed before and after sampling in order to determine total PM$_{10}$ collection, and monosaccharides quantified by using a previously validated GC-MS method (Cordell et al., 2014). The highest concentrations of Lev was found in November and December (126, 95 ng/m$^3$, respectively), the lowest average concentration of Lev was observed in summer (20 ng/m$^3$). The correlation between Lev and PNSD and BC will be investigated. The influence of wind speed and direction on the Lev concentration will also be discussed.
1.018 THE MUMBA CAMPAIGN: MEASUREMENTS OF URBAN, MARINE AND BIOGENIC AIR.

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Abstract:

The Measurements of Urban, Marine and Biogenic Air (MUMBA) campaign took place in Wollongong, New South Wales (a small city approximately 80 km south of Sydney, Australia), from 21st December 2012 to 15th February 2013. Instruments were deployed during MUMBA to measure the gaseous and aerosol composition of the atmosphere with the aim of providing a detailed characterisation of the complex environment of the ocean/forest/urban interface that could be used to test the skill of atmospheric models. Gases measured included ozone, oxides of nitrogen, carbon monoxide, carbon dioxide, methane and many of the most abundant volatile organic compounds. Aerosol characterisation included total particle counts above 3 nm, total cloud condensation nuclei counts; mass concentration, number concentration size distribution, aerosol chemical analyses and elemental analysis. In this paper an overview of the campaign and its findings for the urban, marine and biogenic influences are presented, along with
comparison to regional scale chemical transport model predictions from CHIMERE [Menut, et al, 2013].
1.022 Dependence of Simulated Tropospheric Ozone Trends on Uncertainties in U.S. Vehicle Emissions.

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Abstract:
Long-term surface observations show a rapid increase in background concentrations of ozone since the 1960s. Global chemistry-climate models have difficulties in reproducing the magnitude of this trend, overestimating the mid-century observed concentrations. This suggests that the impacts of ozone on climate and air quality throughout the second half of the 20th century may be misrepresented in current models.

We use the MACCity emissions inventory constrained by ambient observations to examine the dependence of simulated long-term ozone trends on U.S. land transportation (vehicle) emissions of nitrogen oxides (NO\(_X\)), carbon monoxide (CO) and volatile organic compounds (VOCs). Sensitivity simulations are performed using the Community Earth System Model (CESM), where the U.S. MACCity vehicle emissions of either NO or CO, and co-emitted VOCs, are constrained to the observed NO:CO ratio between 1960-2010. We will present results from these sensitivity simulations showing that changes in the VOC and CO emissions lead to improved simulations of ozone in LA and over the U.S. However, we will also show that the impacts of these emission changes are limited outside of the U.S boundary layer.
1.024 Photochemical smog modeling for ozone air quality management in Bangkok Metropolitan Region.

Early Career Scientist

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Abstract:

Surface ozone pollution remains an issue in the Bangkok Metropolitan Region (BMR), Thailand. High emissions of precursors emissions coupled with favorable meteorological conditions create ozone pollution episodes throughout the year, especially in the dry season. The annual average concentration of ozone in BMR is observed with an increasing trend. To understand its formation in order to formulate mitigation strategies for improving the ozone air quality in the BMR, a collaborative research aiming at applications of photochemical smog models was conducted jointly by the Asian Institute of Technology (AIT) and PTT Public Company Limited. A high resolution emission inventory (EI) data was prepared for major anthropogenic sources of on-road transport, industrial, power plant, livestock, farm machine, navigation, locomotive, and residential combustion. Volatile organic compounds (VOCs) emissions from fuel station and biogenic VOC were also included in the EI. Gridded hourly emissions were prepared in 2x2 km² grid resolution with the necessary VOC speciation for modeling input. CAMx/MM5 modeling system was selected to simulate ozone in March and August 2010 which respectively represent high and low ozone pollution month in the BMR domain. Model performance evaluation was conducted for both modeled meteorology and ozone levels using the existing observation which showed acceptable model performances in terms of statistical criteria. The maximum monthly average in the domain of modeled ozone concentrations in March and August were about 41 ppb and 38 ppb while the maximum modeled hourly ozone concentrations were 159 ppb (March) and 158 ppb (August). Ozone formation regime in BMR was more sensitive to VOC than to NOx emission reduction hence to meet the hourly standard of 100 ppb, either a VOC reduction of >50% or NOx reduction of >90% would be required. Various emission scenarios can be analyzed using the evaluated modeling system to propose appropriate emission control strategies.
Modeling of the anthropogenic heat flux and its effect on air quality over the Yangtze River Delta region, China.

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Abstract:

Anthropogenic heat (AH) can affect the city environment. The spatial distribution of AH fluxes in the Yangtze River Delta (YRD) region is estimated, and is also incorporated into the modified WRF/Chem with the seasonal and the diurnal variation. The impacts of accounting for AH fluxes on the meteorology and air quality over the YRD region are studied. The results show that AH fluxes over YRD has been growing from 1990 to 2010. In 2010, the high values of AH over the urban areas of Shanghai can reach 113.5 W/m². Including AH can significantly change the urban heat island and urban-breeze circulations in the cities of the YRD region. In Shanghai, 2-m air temperature increases by 1.6 °C in January and 1.4 °C in July, the planetary boundary layer height rises up by 140m in January and 160m in July, and 10-m wind speed is enhanced by 0.7 m/s in January and 0.5 m/s in July. And the enhanced vertical movement can transport more moisture to higher levels, which causes the decrease of water vapor at the ground level and the increase in the upper PBL, and thereby induces the accumulative precipitation to increase by 15-30% over the megacities in July. The adding AH can impact the simulated pollutants as well. The concentrations of primary air pollutants decrease near surface and increase at the upper levels, due mainly to the increases of PBLH, surface wind speed and upward air vertical movement. But surface O₃ concentrations increase in the urban areas, with maximum changes of 2.5ppb in January and 4 ppb in July. Chemical direct and indirect effects can play a significant role in O₃ changes. AH should be considered in any climate and air quality assessment.
Airborne Observations of Water Vapor Stable Isotope Ratios in the Lower Troposphere around Washington, D.C. and Indianapolis.

Early Career Scientist

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Abstract:

Water vapor is the most important greenhouse gas, contributing about 2/3 to the natural atmospheric greenhouse effect. Many studies provide evidence supporting the positive feedback between increasing global temperatures and atmospheric water vapor concentrations. In addition to being major sources of greenhouse gases contributing to warming, cities have also been shown to modify humidity levels, and influence the frequency and intensity of precipitation events due to differences in land cover and emissions relative to rural areas. Understanding the complex modifications urban areas can have on water cycling is important, as 54% of the world’s population reside in cities, with this number projected to grow over the coming decades.

To investigate processes influencing atmospheric moisture levels around urban areas, such as evapotranspiration, tropospheric entrainment, and anthropogenic water vapor emissions from combustion and evaporative cooling, airborne measurements of water vapor isotopologues containing $^2$H, $^{18}$O, and $^{17}$O were conducted in the boundary layer and free troposphere around the cities of Indianapolis and Washington, D.C.-Baltimore. These flights were prompted by periodic observations of elevated water vapor concentrations in the urban outflow of Washington, D.C.-Baltimore and Indianapolis since 2012. Airborne water vapor isotopologue measurements were conducted during February and March of 2016 using Purdue University’s Airborne Laboratory for Atmospheric Research (ALAR) and an LGR water vapor isotope analyzer. Flights were designed to investigate the isotopic signature of water vapor above rural, suburban, and urban land cover, and included sampling of water vapor point sources, such as plumes from power plants and evaporative cooling towers. Additionally, vertical profiles extending several hundred feet above the boundary layer were conducted upwind and downwind of the urban areas to investigate entrainment effects on the isotopic signature of boundary layer water vapor. Results of the 2016 flights will be discussed.
1.030 Airborne Particles in the Urban-Marine Environment

Early Career Scientist

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Melita Keywood, CSIRO Ocean and Atmospheric flagship

Abstract:

Atmospheric particles were measured in the seaside city of Wollongong, Australia, during an intensive field campaign known as MUMBA (Measurement of Urban, Marine and Biogenic Air) between 21st Dec 2012 and 15th Feb 2013. A scanning mobility particle sizer (SMPS) and eSampler were operated to measure number particle size distributions ranging from 14 nm to 660 nm in diameter and particle mass (PM$_{2.5}$) respectively. Principal component analysis has been applied on the SMPS dataset and revealed three different factors (Large, Medium and Small). The three factors are ranging from 14 nm to 478 nm in diameter and describe 85% of the dataset cumulative variance. All three particle factors and particle mass increased steadily in the morning with a distinct peak observed between 7:00 and 8:00 (Australian Eastern Daylight Saving Time), related to a combination of dispersion of accumulated particles overnight and local traffic. North-westerly winds carry biogenic emissions from the Illawarra escarpment conservation area. Anthropogenic emissions from the central business district and the steel works are identifiable in southerly winds. Total particle concentrations appear to be strongly influenced by the prevailing easterly to north easterly sea breezes that carry pollutants from sources in and around Sydney.
1.031 Factors contributing to the haze pollution in Wuhan during October 2014: local Emissions, regional transport and biomass burning.

Early Career Scientist

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Abstract:

Wuhan as the center of city clusters in central China has frequently experienced severe haze pollution just as Hebei-Beijing-Tianjin, Yangtze River Delta, but the major pollutants sources and transport processes in the developing region remains unclear. We investigated the sources and formation mechanism of the two typical haze processes occurred on October 7-21, 2014 in Wuhan using the Nested Air Quality Prediction Model System (NAQPMS/IAP, China) with source-tagging analysis and sensitivity analysis. This study quantitatively estimated the contribution from the local emissions over Wuhan, the remote emissions from surrounding areas and the biomass burning sources to the PM$_{2.5}$ concentration in Wuhan. The main results are as following: (1) Under the condition of strong northern and northeastern wind and high planetary boundary layer height, the air pollutants could be transported from the severely polluted North China to Wuhan, which formed a short-time haze. The long-range regional transport from the north areas (60% contribution to PM$_{2.5}$ concentration of Wuhan) and the local emission (24%) exhibited strongly influence on PM$_{2.5}$ pollution. Henan province, was found as a key source region that contributed 47% of PM$_{2.5}$ concentration. (2) During the second episode, stagnant atmosphere led to accumulation of air pollutants and then the prolonged haze process in Wuhan. The close-range transport from the eastern and northeastern surrounding regions (25%) and the local contribution (40%), were the dominating sources to PM$_{2.5}$ concentration over Wuhan. Another important source regions identified was Anhui province (daily
contribution reached 28%). (3) The hourly contribution from biomass burning to the PM$_{2.5}$ concentration in the suburb of Wuhan could be 40-60% at certain time, but biomass burning exerted less significant influence on Wuhan’s PM$_{2.5}$ pollution during the entire haze period. Our results indicate that reducing the local emissions over Wuhan would be not sufficient to control the PM$_{2.5}$ pollution of Wuhan.
1.034 The Global Reactive Carbon Budget of the Troposphere: a Modeling Perspective.

Early Career Scientist

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Abstract:

Reactive carbon plays a key role in driving the oxidative chemistry of the atmosphere and is a precursor to ozone and particulate matter formation, both of which impact human health and climate. However, uncertainties on the lifecycle of reactive carbon in the atmosphere remain large. In part this can be attributed to the chemical complexity and variety of species reactive carbon in the atmosphere, and the resulting lack of mass closure in both ambient measurements and models. Furthermore, the complex oxidative chemistry of the troposphere and the implications for the formation and loss of organic aerosol, encourage a holistic perspective on the sources and sinks of all the reactive carbon in the atmosphere.

In this study, we present the first attempt to simulate the reactive carbon budget of the troposphere using a global chemistry transport model, GEOS-Chem. We add the oxidation of aromatics and monoterpenes to the existing chemical mechanism, we expand the treatment of dry and wet removal of organic species, we introduce a complete mass tracking of all reactive carbon species, and finally we update the chemical mechanism to achieve carbon closure. We use this updated model to construct a global reactive carbon budget, comparing the relative magnitude of the sources (emissions, oxidation) and sinks (wet removal, dry removal, oxidation to CO$_2$) of atmospheric reactive carbon. This study presents a new perspective on the processes controlling the spatial and temporal distribution of anthropogenic and biogenic reactive carbon in the atmosphere.
Monitoring and Forecasting Air Quality over China: Results from the PANDA Modeling System.

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Abstract:
With fast economic growth and development China is experiencing severe air pollution episodes related to rapid industrialization and urbanization since more than three decades. Through collaboration between 7 European and 7 Chinese research universities and institutes, the PANDA (Partnership with China on Space Data) EU-funded project aims to improve our understanding of the processes responsible for the formation, dispersion and destruction of air pollutants in East Asia. By combining space and in-situ observations and surface emissions of chemical pollutants with global and regional models of atmospheric composition, detailed analyses and reliable forecasts of regional air quality are produced with the aim of improving methods for monitoring and forecasting air quality in East Asia.

Using a multi-model approach based on several global and regional state-of the art model simulations, we present detailed modeling studies of recent notorious haze events in East Asia (winter 2010 and 2013 haze events). We will demonstrate the effect of uncertainties and differences in current emission inventories for China and their resolution and temporal variation on model performance. We will focus especially on major city clusters that have experienced intense urbanization and population growth. The importance of using a downscaling approach to better reproduce and predict air pollution events that occur in East Asia will be discussed. The relation between the performance of the models and the complexity of their chemistry and aerosol schemes and choice of boundary and atmospheric forcing will be assessed. We will also present results of the air quality forecasting system being developed within PANDA and in collaboration with the EU project MarcoPolo.
1.036 Evaluation of the MOSAIC air quality box model using recent field campaign observations.

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Abstract:

Future air quality will be influenced by various global change factors such as climate, US anthropogenic emissions, wildfire emissions, biogenic emissions, land cover, and background concentration changes associated with long-range transport. Understanding how each global change factor contributes to the changes in future air quality is of interest. However, given the fact each factor has a range of future projections, ensemble analysis of these global factors using three-dimensional chemistry-climate or chemistry transport models is very computationally burdensome.

MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) is an atmospheric chemistry box model with a full and explicit treatment of gas and aerosol phase chemistry and dynamics. Here, we develop an approach using MOSAIC as the basis for an assessment of global change factors for key air quality issues over the Western US, including wintertime stagnation events, summertime urban to rural transport cases, and wildfire events. In this presentation, we will present results from an initial evaluation of the MOSAIC model using observations from recent field campaigns. In particular, we will focus on the model evaluation against the CARES field campaign (Carbonaceous Aerosols and Radiative Effects Study), which was carried out in June 2010 in the central California region. This field campaign included a comprehensive list of gas, aerosol, and meteorological measurements of the Sacramento urban plumes as they were transported into the forested Sierra Nevada areas, making it a good test case for the application of the MOSAIC Lagrangian air quality box model.
1.038 Isotopic constraints on sulfate and nitrate aerosol formation mechanisms in Chinese haze events.

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Abstract:

An estimated 1.6 million people die prematurely annually in China due to severe air pollution. Particulate matter smaller than 2.5 microns in diameter (PM$_{2.5}$) is the worst offender, and concentrations can reach upwards of 500 mg m$^{-3}$. Observations have shown that secondary inorganic aerosols (sulfate, nitrate, ammonia) are a large component PM$_{2.5}$ mass, and that their mass fractions increase substantially during haze events, suggesting increased heterogeneous formation rates. Models have been unable to simulate the high concentrations of PM$_{2.5}$ and sulfate that are observed, and this low model bias has been attributed to a missing chemical source of sulfate aerosol production in models. We present observations of the oxygen isotopic composition of sulfate and nitrate aerosol from samples collected in fall and winter in Beijing. The oxygen isotopes of secondary sulfate and nitrate aerosol are determined by their chemical formation pathways, and thus provide an observational constraint on the unique chemistry occurring in this megacity. We use a global model (GEOS-Chem) with a high-resolution nested grid over China, which contains the oxygen isotope tracers, to quantitatively interpret the isotope observations. We will provide the first observation-based estimate of the dominant formation mechanisms of sulfate and nitrate aerosol during Beijing haze events, and insight into the missing source of sulfate in air quality models.
1.039 Influence of NOx emissions on Central Valley fog frequency and persistence.

Early Career Scientist

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Abstract:

From 1930-1970, California’s Central Valley (CV) radiation fog significantly increased, with locations such as Fresno seeing an ~83% growth in fog frequency. However, in the last 30 years, researchers identified a ~50% reduction in fog days (Baldocchi and Waller, 2014, Herkes et al., 2014). The dominant hypotheses suggest that the decline in fog can be explained by rising temperatures associated with climate change or urban heat island effect. This assertion fails to explain the significant increase in CV fog midcentury. Here we assert that changes in air pollution, rather than climate, better explain this upward-then-downward temporal trend. Growth in vehicle use greatly increased emissions of NO_x midcentury, followed by a significant decrease in air pollution due to statewide regulations from 1980-Present. In the CV, ammonium nitrate (NH_4NO_3), the dominant wintertime aerosol, is limited by NO_x concentration. NH_4NO_3’s size range and hygroscopicity make it an important source of cloud condensation nuclei (CCN). Thus, air pollution growth from 1930-1970 increased the availability of CCN necessary for fog formation. Subsequent air pollution mitigation after 1980 then reduced NO_x, and thus CCN and fog frequency.

Using over 75 years of meteorological measurements, we developed a detailed fog climatology spanning 15 locations. Additionally, we developed a historical record of nitrogen oxide (NO_x) throughout these sites to determine air pollution trends. We used this data to analyze the spatial and temporal correlation between fog frequency, air pollution, and climatic drivers.

CV fog exhibits a pronounced north-south gradient, with fog consistently more frequent and persistent in southern latitudes than northern. Additionally, NO_x concentration also shows a similar north-south gradient, with concentration consistently highest in the south and a steady ~50% decline in all sites since 1990. We conclude that fog trends in the CV best correlate both temporally and spatially with NO_x trends, rather than with climatic drivers.

Abstract:
Combustion and its associated emissions are important consequences of urbanization. However, accurate monitoring and quantitative assessment of these consequences, especially in rapidly developing regions, are hindered by the lack of information regarding combustion activities and efficiencies. There is a unique opportunity to augment our observational capability by using multiple datasets from ground, aircraft, and satellite products as well as emission databases and chemical transport models (CTM) and associated reanalysis. In particular, joint analysis of CO, CO\(_2\) and NO\(_X\) which are co-emitted provide a unique perspective to study anthropogenic combustion given the availability of satellite retrievals for these combustion products. Here, we analyze these three species, including their associated ratios and trends using satellite observations (Measurement of Pollution In The Troposphere, Ozone Monitoring Instrument, Tropospheric Emission Spectrometer and Greenhouse gases Observing Satellite, Orbiting Carbon Observatory), available reanalyses, and emissions from EDGAR and RCPs in all major cities in US and applying similar methodology to all megacities around the globe. We also compare results from various data sources (ground and aircraft measurements) for verification of our trend analysis. Temporal evolution within one city and comparison between cities provide knowledge on how the city and its natural/artificial environment interact. Our initial results show that: 1) There are obvious differences in trends between satellite-based and emission-based data. 2) Distinct patterns are found in bulk characteristics in regions with clear fire or fossil fuel combustion. 3) Generally, CO/NOx decreases with time and this ratio is lower in developed countries than developing countries with the exception of rapidly urbanizing regions in China where both CO and NOx are significantly high. We will couple our results with socio-economic indicators of urbanization such as population, GDP, economic structure and development patterns, to provide an integrated perspective to city evolution and interaction between urbanization, anthropogenic activity, and our environment.
Towards an International Network for Monitoring, Analyzing and Forecasting Regional Air Quality.

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Abstract:

Urbanization with its concentration of population, economic activities industrial development and business has generated an increase in the atmospheric level of air pollutants with adverse effects on human health, food production and well-being. Addressing this major problem, specifically in urban areas of Asia, Latin America and Africa, requires the development of coherent measurement, analysis modeling and dissemination tools with the highest possible standards. We propose to develop and implement a global interdisciplinary network of experts in different parts of the world who will work together and develop state-of-the-art regional air quality prediction systems with downscaling and evaluation capability from the global to the regional and local scales. We invite IGAC and other international programs to sponsor and support this worldwide initiative.
Investigation of Secondary Organic Aerosol (SOA) formation pathways in Beijing using an observation-based SOA model.

Early Career Scientist

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Abstract:

SOA has been underestimated over China by current modeling studies to a larger degree than over Europe and the USA. In this study, we investigated SOA formation pathways in urban Beijing using a box model constrained by observations. Our model considers the multi-generation oxidation process of volatile organic compounds (VOCs) and intermediate VOC (IVOCs), and the chemical aging of semi-volatile primary organic aerosol (POA). Our results showed that the SOA formation rate reached 30.3 μg m⁻³ day⁻¹ in Beijing over the investigated two-day period. SOA produced from VOCs, IVOCs and POA contributed 14%, 82% and 4%, respectively. Contribution from IVOCs was similar to the magnitude of previous model underestimation which has been conducted. Considering the uncertainties in the emission rates and the POA volatile distribution factors, SOA yield in Beijing was 80.0 μg m⁻³ with a range of 60.0-102.6 μg m⁻³. We found that the oxidation of IVOCs contributed to the majority of SOA production in Beijing during the study period, which was not considered in previous simulations in China, and which likely improves the three-dimensional model SOA predictions in China. The SOA formed by IVOCs remains a large source of uncertainty, and the lack of observations in China make it difficult to choose the correct modeling parameters. The emission sources and reaction rates of IVOCs are needed for further identification and quantification purposes.
1.048 Source apportionment modelling of OC and NMVOCs in the Berlin urban area.

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Abstract:
A 3-month measurement campaign (BAERLIN-2014) was carried out during the summer of 2014 in the urban area of Berlin and Potsdam, Germany. A variety of gas-phase, particulate matter (PM), and meteorology parameters were measured. To investigate source contributions, including the role of anthropogenic vs biogenic influence in the urban area, various source apportionment techniques were applied. Chemical mass balance source apportionment was utilized for PM10 filter samples, specifically the organic carbon (OC) fraction. Higher time-resolution NMVOC data was collected by PTR-MS and used in positive matrix factorization source apportionment analysis. Initial results indicate a significant influence of vegetation as well as traffic for both the OC and NMVOC source apportionment results. In addition, the largest contribution to OC was from secondary organic aerosol (SOA). The OC source apportionment analyses were complemented with air mass back trajectory data and bulk PM composition analysis, including inorganic contributions from sulfate, nitrate, and ammonia, which were significant contributions to the PM mass. Furthermore, the source apportionment results were compared to the existing understanding of sources based on local emission inventories, finding that in the case of NMVOCs, the attribution in the emission inventory to solvents was greatly overestimated relative to the importance of traffic sources. These results are similar to findings comparing measurements to emission inventories for other cities in Germany and Europe.
1.050 Improving our understanding air quality satellite measurements: an update from KORUS-AQ.

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Abstract:

The atmospheric composition constellation (ACC) of air quality measurement satellites is currently being developed. The ACC will have global coverage provided by the ESA sun-synchronous low earth orbit Sentinel-5 Precursor (TROPOMI) satellite. In addition, three geostationary satellites in the northern hemisphere will provide complementary hourly daytime measurements for much of Asia (NIER GEMS), North America (NASA TEMPO), and Europe (ESA Sentinel-4). As the ACC comes closer to existence the international atmospheric composition community needs to develop a coordinated plan to execute field campaigns and build up sufficient networks of ground-based insitu and remote sensing instruments designed to improve our understanding of what the space-borne air quality sensors are measuring. In addition we need to these air quality satellites how these air quality satellites can provide the various air quality stakeholders the value added data products they need to address their science and policy questions. This presentation will be a first look at the preliminary results from May-June 2016 KORUS-AQ field campaign in South Korea with the goal of describing any new insights related to the challenges of extracting surface concentrations of O₃, NO₂, HCHO, and PM2.5 from column measurements of air quality as measured by UV/VIS spectroscopic sensors.

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Abstract:

Since 1980, global emissions of ozone precursors have shifted toward the equator as less industrialized nations have increased their emissions and industrialized mid-latitude nations have reduced emissions. Modeling studies have also shown that the global tropospheric ozone burden is much more sensitive to changes in emissions in the tropics than in the mid-latitudes. However, previous research has not previously quantified the importance of the historical change in emission location on the tropospheric ozone burden. Here we use global model simulations to separate changes in the global tropospheric ozone burden from 1980 to 2010 into components due to changes in: i.) the global spatial distribution of emissions, ii.) the total magnitude of global emissions, and iii.) the global methane concentration. Using the CAM-chem global atmospheric model, we show that the change in the spatial distribution of emissions is responsible for over half of the net ozone burden change from 1980 to 2010 – more than the effects of the global emission magnitude and the methane change combined. This greater sensitivity to tropical emissions is caused by the strong convection, fast photochemistry, and highly NOx-sensitive conditions in this region, despite the shorter lifetime of ozone. We present evidence suggesting that emissions increases from Southeast Asia, East Asia, and South Asia have been most important for this global ozone increase. The spatial distribution of emissions, particularly emissions from the tropics, dominate global ozone. Previous analyses may have put too much emphasis on emission magnitude and too little on emission location. For example, a continued equatorward shifting of emissions may cause global ozone to continue to increase even if global ozone precursor emissions decrease.
1.058 Quantifying uncertainties in multi-pollutants health impacts in urban/rural regions across the UK.

Early Career Scientist

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Abstract:

The adverse impacts of air pollution on human health due to exposure to ozone (O$_3$), and PM$_{2.5}$ are well established. However, there are numerous uncertainties in quantifying region-wide health impacts e.g. due to uncertainties in simulating urban air pollutant concentrations and for coefficients that determine exposure-response relationships. Previous studies using chemical transport model simulations outline the importance of model resolution for simulating O$_3$ concentrations to be used in health impacts studies. However there are very few studies that discuss the effects of uncertainty in model resolution for determining PM-related health impacts. In this study we have used the UK chemistry and aerosol (UKCA) model to quantify the impact of model resolution and uncertainty in concentration-response coefficients on simulated pollutant concentrations and associated health impacts.

Two model configurations were used: a global resolution (~ 150 km) and a regional resolution (~ 50 km) over Europe. Our regional configuration shows similar results to previous studies for O$_3$ concentrations, in particular better agreement with measurements for the diurnal cycle for O$_3$ compared to global model results. In addition, the regional simulation better captures the lower O$_3$ levels associated with high NO$_X$ levels in large cities (due to higher emission resolution). In contrast, differences in model performance for PM$_{2.5}$ for the two resolutions are not as evident. However the regional configuration gives a better representation of hotspots whereas the global configuration underestimates high PM$_{2.5}$ levels. These results are being linked to population and baseline mortality data to predict uncertainty ranges for PM$_{2.5}$ health impacts over Europe due to long-term exposure.

UKCA simulations at the local scale (~ 12 km) will also be utilised to assess health impacts due to PM$_{2.5}$ episodes across the UK. Future work will consider how health burdens will change in urban areas due to higher population density and climate change.
1.060 Nitrogen oxides and specified hydrocarbons – twenty years of ambient observations in air masses dominated by traffic emissions in Germany.

Early Career Scientist

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Abstract:

During the past 20 years the number of violations of ozone alarm threshold values decreased to nearly zero in Germany. This air quality improvement was achieved through the reduction of traffic related emissions in urban areas. We analyse ambient observations of nitrogen oxide and hydrocarbon (VOC) concentrations. Diesel vehicles can be identified as the dominating source of nitrogen oxide emissions. Detailed hydrocarbon composition measurements identify temporarily reduced catalyst efficiency of petrol cars as the major source of VOC emissions in urban areas. Taking the reactivity of the individual hydrocarbons with the OH radical into account, the total VOC reactivity (R\text{VOC} = \sum \tau k_{\text{OH}+\text{VOC}} \cdot [\text{VOC}] ) can be calculated from the measured VOC concentrations. Together with the reactivity of nitrogen oxide with the OH radical (R\text{NO}_2) this parameter can be used to describe the local ozone production instead of using the full details of the hydrocarbon mixture.

Our observations show a decrease in the R\text{VOC}/R\text{NO}_2 ratio by a factor 7.5 during the period from 1994 to 2014. The analysis revealed that the reduction of the local ozone production rate was mainly caused by large VOC reduction combined with small NO\textsubscript{x} reduction at the same time.

In terms of air quality, future effects of new technologies for nitrogen oxide reductions for diesel cars will be discussed as well as a discrepancy between the observed VOC composition and the emission inventories: While the traffic related VOC emissions have
been significantly reduced in line with emission inventories, the inventory reports approximately constant VOC emissions from solvent use, being the dominating source of VOC in Germany. This is in contrast to our observations in cities where solvent use seems to be a minor VOC source. The contributions of the different sources will be discussed, based on ongoing measurements ranging from urban to remote regions.
1.064 Urban pollution to the Andean cryosphere.

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Abstract:

Large portions of South America’s population live west of the Andes as well as in the Andean valleys and plateaus, with a strong dependence on water resources from the melting of Andean glaciers and snow. Various studies have shown that these glaciers have receded in the last decades in response to temperature and precipitation changes. However, absorbing particles such as black carbon (BC) and dust could potentially also contribute to the increase of melting rates. Important sources of these pollutants, such as cities and mining activity, are located on the slopes or vicinity of the mountain range. The study of the potential transport of urban pollutants to the Andean cryosphere is the focus of several projects and initiatives that have been started in the last few years in the region. Preliminary results of campaigns measuring transport up into the Andes as well as model outputs simulating this transport will be presented.
1.066 Does vehicular use ban program will improve air quality in Mexico Megacity?.

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Abstract:

During March, 2016 the pre and contingency programs were applied in Mexico City, those programs for controlling air quality were no executed since 2002 for ozone. After that a new program for controlling emissions were published. One the measure considers to forbid drive for a 20% of the vehicular fleet per day including Saturdays. In this work an analysis of air pollutants, emissions inventories, information from traffic and air quality modeling is presented in order to evaluate the effectiveness of such action on the ozone reduction in Mexico Megacity
Nitryl chloride as a ‘new’ radical source and its impact on ozone in polluted troposphere: an overview of field measurement and model results in China.

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Abstract:

Nitryl chloride (ClNO₂), which is produced from heterogeneous reactions of dinitrogen pentoxide (N₂O₅) on chlorine containing aerosols, can significantly affect radical budget and concentrations of ozone and other secondary pollutants. However, the abundance, formation kinetics, and impact of ClNO₂ are not fully understood in different environmental conditions. This poster presents an overview of recent field campaigns of ClNO₂ and related chemical constituents in China, including one at a mountain top (957 m a.s.l) in Hong Kong of South China in winter 2013 and three in North China (urban Ji’nan, semi-rural Wangdu, and Mt Tai (1534 m a.s.l)) in summer 2014. ClNO₂ and N₂O₅ were measured with a chemical ionization mass spectrometer (CIMS) with iodide as the primary ions. Ambient concentrations of several hundreds ppt and up to 4.7 ppbv of ClNO₂ were observed in these locations, suggesting existence of elevated ClNO₂ in both coastal and inland atmospheres of China. Measurements in North China exhibited generally low concentrations of N₂O₅, indicative of its fast uptake on aerosols under aerosol and humid conditions. Indications of anthropogenic sources of chloride were observed at all these sites. The impact of photolysis of ClNO₂ on radical budget and ozone was assessed with a MCM model which was updated with detailed chlorine chemistry and constrained by measurement data. The results show that the ClNO₂ could increase ozone production by 2-16% in the following day. An improved WRF-Chem chemical transport model was applied to Hong Kong-Pearl River Delta region to simulate the spatial impact of N₂O₅ uptake and Cl activation on ozone and reactive nitrogen. Overall, our study re-affirms the need to include ClNO₂ related reactions in photochemical models for prediction of ground-level ozone and secondary aerosols in polluted environments.
Improving the short-term air quality predictions over the U.S. using chemical data assimilation and analog-based uncertainty.

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Abstract:

The National Oceanic and Atmospheric Administration National Air Quality Forecasting Capability (NAQFC) is one of the key tools used by decision makers across the U.S. to protect the public from poor air quality. This project funded by the National Aeronautics and Space Administration (NASA) aims to enhance the decision making activity by improving the accuracy of NAQFC short-term predictions of ground-level ozone and particulate matter less than 2.5 µm in diameter (PM$_{2.5}$) and to provide reliable quantification of their uncertainty, by exploiting NASA Earth Science Data with chemical data assimilation and analog-based approaches. The project has following three main objectives:

- The first objective is to improve the initialization of the NAQFC operational air quality prediction system, which is based on the Community Multiscale Air Quality (CMAQ) model, via chemical data assimilation of multiple satellite retrieval products within the community Gridpoint Statistical Interpolation system. We are currently developing a framework in GSI to assimilate retrievals of aerosol optical depth from the NASA Aqua/Terra Moderate Resolution Imaging Spectroradiometer, carbon monoxide from the Measurement of Pollution in the Troposphere and nitrogen dioxide from the Ozone Monitoring Instrument. We also plan to assimilate surface observations of PM$_{2.5}$ (and possibly of ground-level ozone) from selected stations of the AIRNow and the Interagency Monitoring of Protected Visual Environments.
- The second objective is to improve the CMAQ deterministic predictions considerably and reliably quantify their uncertainty with a probabilistic analog ensemble (AnEn) applied to the CMAQ deterministic predictions.
- The third objective is to extrapolate the deterministic and probabilistic point-based...
predictions to a two-dimensional grid over the U.S.

This presentation will report results from each of the aforementioned project objectives, quantify the developed system performance with a range of metrics for both deterministic and probabilistic PM$_{2.5}$ predictions across several hundreds stations over the U.S.
1.078 Supporting decision-making processes through the analysis of air quality policies acceptability.

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Abstract:

The SEFIRA (Socio Economic Implications For Individual Responses to Air Pollution Policies in EU27) EU FP7 project has been conceived in order to support the review and implementation of the air quality legislation, improving its effectiveness and acceptability. This task has been achieved through the coordination of trans-disciplinary scientific and socio-economic resources. Air quality policies are not implemented in a social vacuum; they require a continuous interaction with individuals, often implying significant changes in their lifestyles. In addition, the extent to which people endorse a policy is crucial in determining its effectiveness and success both at national and regional scales. However, while there is a broad consensus that public acceptability affects both the effectiveness and success of environmental policies, acceptability has not been fully studied and internalised in the models used to support the policies' adoption. Exploring different quality and quantitative approaches, we found that discrete choice analysis might offer a comparative advantage in supporting the decision-making process in particular when the implementation process involves behavioural changes. Discrete Choice Experiments (DCEs) allow investigating people’s preferences and their potential behaviour, identifying variables affecting individual choices between two or more “choice alternatives”. Within SEFIRA a choice experiment has been conducted, based on 16.100 interviews administered in seven European countries. In this experiment the social aspects related to individual choices have been taken into considerations and socio-economic data of respondents have been used, allowing us to perform a segmentation analysis and highlighting socio-economic differences in the air quality acceptability across the various countries. The results of our analysis suggest that the willingness of citizens to change their behaviour might be larger than previously estimated. In addition, differences in policy acceptability by country and socio-economic structure of the population are highlighted.
Evaluation of performance of simulated secondary pollutants by using air quality models for urban areas in Japan.

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Abstract:
Emissions and concentrations of primary atmospheric pollutants (eg. NOx and VOCs) have been decreased on urban areas in Japan owing to conduct their emission regulations. Nevertheless, PM2.5 and photochemical oxidants (Ox) have still remained high concentration levels and their air quality standards of Japan have not been attained at most monitoring stations (Ministry of the Environment of Japan (MOE), 2015). For forming effective air pollution control strategies, currently, we raise expectations for applying air quality models reproducing complicated physical and chemical processes of both of primary and secondary pollutants.

The urban air quality model inter comparison study in Japan (UMICS) was started to improve performances of air quality models (eg. Chatani et al., 2014). UMICS showed some critical problems immanent in the air quality models. For example, the models tended to overestimate NO$_3^-$ but to underestimate OA, although simulated PM2.5 concentrations were reasonable with comparing to observations at Kanto area (Shimadera et al., 2014). In terms of O$_3$, the models reproduced well the diurnal and inter-diurnal variations in the O$_3$ concentrations at most observational stations in Kanto area but tended to overestimate nighttime O$_3$ and to underestimate daytime O$_3$ at several observational stations (Morino et al., 2010).

In order to find the causes of discrepancies between the simulated and observed concentrations of PM2.5 and O$_3$, in this study, air quality simulations were performed under different model settings by using the Weather Research and Forecasting (WRF) model and the Community Multi-scale Air Quality (CMAQ) model system. Performances of simulated secondary pollutants using these model settings were evaluated by comparing with observed concentrations of secondary pollutants (O$_3$, PM2.5, and constituents of PM2.5) on urban areas in Japan, which were provided by UMICS and MOE.
1.083 Analysis of first PM2.5 samples from a Harvard Impactor located in the city of Natal, Brazil.

Early Career Scientist

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Abstract:

We present a chemical characterization of PM2.5 aerosols collected by a Harvard Impactor in the metropolitan region of Natal, Brazil. Samples were taken on a daily basis using 37 mm quartz filters, which were then analyzed by gravimetric techniques, quantification of Black Carbon (BC), and fluorescence X-ray analysis. The PM2.5 sampling occurred continuously during the period of March 3, 2015 to March 31, 2016. First analyses from March to August 2015 show that PM2.5 monthly averages ranged from 3.38-14.97 μg/m³, while daily means ranged from 0.25-70.98 μg/m³. The primary daily PM2.5 standard of 25 μg/m³ set by the World Health Organization is exceeded for nine times in this period, and the daily standard of 35 μg/m³ established by the U.S. Environmental Protection Agency is exceeded more than six times. The month of June showed the highest monthly mean BC concentrations, reaching 1.18μg/m³ as well as the lowest PM2.5 monthly mean concentrations. Highest monthly mean concentrations of PM2.5 were reached in March. Meteorological conditions in June are comparable to those of March, as during both months it rained on average 300 mm. However, cultural facts enhanced the BC concentrations in June as many open wood burning bonfires occur due to the celebrations of the St. John’s holiday. Analysis by X-ray fluorescence identified the chemical elements of the collected particles with highest concentrations in the filters: S, K, Si, Al, Cl, Fe, P, Ca, Mn, and Ca. Most of these were of anthropogenic origin according to their enrichment factor, while Na, Mg, Sn, Zn, Al, Fe and Ti originated from natural sources. These results for the full year as well as HYSPLIT trajectory model simulations of PM2.5 dispersion and deposition will be presented.
1.088 Ozone variations at and around a high altitude site in the central Himalayas.

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Abstract:
In-situ observations of trace gases are severely lacking in the economically developing countries, particularly in the tropics where photochemistry is intense. These observations are more important and critical in the regions where spatial variations in emission characteristics, environment and topography are large within a small region. Here, we report the extensive observations of surface ozone carried out around a high altitude site in the central Himalayas. Observations are made at Manora Peak (29.4 N, 79.5 E, 1958 m amsl) and at a school in Nainital Town. Both the observation sites are at nearly similar altitude and less than about three km apart. A campaign based observations are also made during different seasons in last 4 years, using a mobile van that started from Manora Peak, moved around surroundings and moved down the mountain region, covering different small villages, towns, densely polluted regions, few rural areas and a small-scale industrial region. Large differences are seen in ozone variations at Manora Peak and Nainital town. Dominance of the photochemical ozone production is observed even at a high altitude Nainital town region, however, simultaneous observations made at Manora Peak do not show such increase. In-contrast, there have been occasions, when daytime ozone decrease is observed at Manora Peak. Contrasting ozone variations are also seen at both the sites during an Indian festival, when large amount of fire crackers are burnt. Observations from mobile van showed daytime ozone decrease by about two times, when moving downward from about 2000 m to about 400 m altitude region. Few incidents are observed with simultaneous enhancement in ozone levels in the urbanized region and the central Himalayas, suggesting transport of ozone rich air-masses to the pristine region. Results will be presented in details during the conference.
1.089 High resolution simulations of black carbon aerosols and their vertical stratification over Santiago and its transport to the Andean cryosphere.

Early Career Scientist

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Abstract:

Santiago, Chile (33.5°S, 70.5°W, 500 m.a.s.l., 7 millions inhabitants) is a large city situated by the high Andes. It is regularly affected by high particle concentrations (up to 50 µg/m$^3$) containing black carbón (BC) associated with mainly traffic (diesel combustion), and wood burning but also agricultural burning and fires in summer and fall. To date, it is unclear how far and how often the urban (and peri-urban) pollution plume reaches the Andean cryosphere. Here we explore the region’s complex atmospheric circulation and particularly the transport of traffic and Wood burning BC using a state of the art numerical model, and an updated emission inventory. We compare results against available observations including vertical BC profiles obtained in measuring campaigns in 2013 and 2015.
The causes and consequences of unanticipated decreases in carbonaceous aerosol over the U.S. between 1990 - 2012.

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Abstract:

Exposure to atmospheric particulate matter (PM) exacerbates respiratory and cardiovascular conditions, resulting in ~3.7 million premature deaths per year globally, and ~200,000 per year in the U.S. alone. There has been a significant decline in PM since 1990 in the U.S., attributed primarily to the reduction in inorganic aerosol following implementation of the Clean Air Act amendments (CAAA). Here, using surface observations between 1990 and 2012, we show that both black carbon and organic carbon have also declined dramatically across the entire U.S. by 25-50%; the latter being unexpected and accounts for over 30% of the U.S.-wide decline in PM. Using a chemical transport model, reanalysis meteorology and a new annually varying anthropogenic emissions database for carbonaceous aerosol, we show that the decrease in carbonaceous aerosol can be explained by changes in anthropogenic emissions. Furthermore, the organic aerosol concentration declined despite opposing trends from natural sources. We assess the population exposure and health impacts related to the associated decline in PM, finding that CAAA controls affecting organic aerosol, primarily from vehicle emissions and fuel burning, are responsible for averting 120,000 (59,000 – 197,000) premature deaths between 1990 and 2012; with approximately 69,000 more lives saved than anticipated by the EPA between 2000 and 2010.
Health risk assessment of toxic heavy metals in fine particles of road dusts in Dhaka city, Bangladesh.

Early Career Scientist

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Abstract:

Street dusts were collected from different road sides (22 locations) in Dhaka city, Bangladesh. Dusts samples were sieved below 90 µm and analyzed with EDXRF for fifteen toxic heavy metals. Concentrations of all elements are above on the safe limit of WHO. Pollution assessment is present by studding enrichment factor(EF),pollution index(PI), pollution load index(PLI), and Geo-accumulation index(Igeo).Spatial distribution of the elements has been shown by ArcGIS software.Correlation, cluster and principal component analyses identifiedprobable natural and anthropogenic sources of contaminants. Enrichment factors of Cu, Pb, Cd and Zn showed that the dust is extremely enriched in these metals. Multivariate statistical analyses revealed that Cu, Pb, Zn, Fe and, to a lesser extent, Cr and Ni have common anthropogenic sources. While Mn and Li were identified to have natural sources, Cd may have different anthropogenic origins. The health risk assessment has studied by USEPA model.Elemental health risk was assessed through dose calculations for carcinogenic andnon-carcinogenic metals; and by the determination of LADD (lifetime average daily dose). The carcinogenic metals studied showedthe following variation in their LADD values; Cr>As>Co>Cd>Ni. Dose calculations for non-cancerous elementsshowed that hazard index (HI) of Cr ˃1 and Cd, V also near to 1.So these elements are on non-carcinogenic risk. Dose calculations for cancerous elementsshowed that Cr and Aslie within the threshold of $10^{-3}$-$10^{-6}$ mgkg$^{-1}$day$^{-1}$. So these two elements are on cancerous risk.In Dhaka, it appears that the highest levels of risk for children than adults by the three exposure pathway(ingestion, inhalation and dermal contact) and that two elements—As and Cr—are of most concern regarding the potential occurrence of health effects.
1.102 Using a mobile laboratory and continue wavelet transform to evaluate vehicular emission in megacity Beijing.

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Abstract:

Vehicular emission is a major source of air pollutants in cities, especially in megacities, around the world. Due to the different energy profiles and developmental stages, the contribution of vehicular emission to the air pollution varies greatly in cities around the world. Beijing municipality government implements strict vehicular emission control policy to improve air quality. However, the policy effectiveness has been controversial. During the Asia-Pacific Economic Cooperation (APEC) Beijing 2014, the government adopted the strictest vehicular emission control policy, which provides a great opportunity to re-examine this issue. We used a mobile research platform to measure the main air pollutants, including PM$_{2.5}$, BC, SO$_2$, CO, NOx and O$_3$ on the 4th ring road of Beijing, combined with a continue wavelet transform method (CWT) to separate out “instantaneous emission” from on-road background concentration. The daytime measurement concentrations (mean±1 SD) of PM$_{2.5}$, BC, SO$_2$, CO, NOx and O$_3$ were 11.8 ± 5.3 μg m$^{-3}$, 1.2 ± 0.7 μg m$^{-3}$, 6.9 ± 4.9 ppb, 1.2 ± 0.6 ppm, 215.1 ± 61.1 ppb and 7.8 ± 5.2 ppb on the 4th ring road during APEC period, respectively. Our results suggested that the CWT method could effectively decompose the “instantaneous concentration” from on-road background concentration. The daytime vehicle emission of CO and NOx decreased by 28.1% and 16.3% during APEC period
relative to before APEC period, and by 39.3% and 38.5% relative to after APEC period. The nighttime vehicle emission of CO and NOx decreased by 56.0% and 60.7% during APEC period relative to after APEC period. Our results suggest vehicle emissions of NOx and CO contribute considerably to the total emission of air pollutants in Beijing, and the vehicle emission control is essential in the air quality improvement in megacity Beijing.
1.103 Volatile Organic Compounds at the Urban, Marine and Biogenic interface in South-East Australia.

Early Career Scientist

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**Abstract:**

Ambient concentrations of VOCs were measured in Wollongong, NSW, as part of a large ambient measurement campaign that took place in summer 2012-2013. The campaign yielded a rich dataset characterising atmospheric composition at the ocean/forest/urban interface, an environment which is typical of many cities in south-east Australia. The 8-week campaign yielded time series for VOCs from which typical ambient summertime values were derived. The most abundant VOCs were all oxygenated species (methanol, formaldehyde, acetone and acetaldehyde). The main source of the measured VOCs was the forested escarpment that encircles the Wollongong region to the west. Comparison with measurements made in 1996-2001 indicates decreased emissions of anthropogenic VOCs from vehicular traffic in the intervening years. Biogenic VOC mole fractions an order of magnitude higher than average were associated with atypically warm weather on two days in January 2013. These elevated levels of VOCs were associated with higher ozone. This has implications for air quality policy under a warming climate, since biogenic emissions cannot be subjected to emission controls.
1.104 Evaluation of VOC emissions from vehicles during parking events in Japan.

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Abstract:

Evaporative emissions from vehicles for Japanese market have been evaluated using VT-SHED. We also evaluated a carbon canister purging activity during driving with a chassis dynamometer. Using these results, we estimated real world VOC emissions during parking events considering a state of carbon canister. VOC emissions increased dramatically when a breakthrough of carbon canister occurred. And we also evaluated the effect of introducing stricter regulation for the evaporative emissions on the VOC emissions in Japan.
Insights into the contribution of residential biomass burning to urban pollution in two Colorado cities.

Early Career Scientist

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Abstract:

Residential biomass burning is a prominent means of heating and cooking in U.S. cities; however, its emission profile is source-dependent and its contribution to urban volatile organic compounds (VOCs) remains uncertain. We report wintertime measurements of domestic burning VOCs from two Colorado cities, Boulder and Aspen, using a newly developed high mass resolution $\text{H}_3\text{O}^+$ chemical ionization mass spectrometer. Mobile measurements of residential biomass burning plumes demonstrated that emissions were dominated by small oxygenates such as methanol, ethanol, and acetone. Compared to other point sources such as vehicle exhaust and solvents, burning emissions exhibited unique enhancements of furan compounds such as furfural, furanone, and methyl furan. Residential burning emissions of nitrogen-containing VOCs such as acetonitrile and acrylonitrile were lower by nearly an order of magnitude when compared to controlled burning of agricultural fuels. Background acetonitrile concentrations in Boulder were high and mobile measurements demonstrated elevated concentrations downwind of commercial and academic laboratories. These point sources likely resulted from the use
of acetonitrile as a chemical solvent; consequently, acetonitrile may not be a suitable tracer for residential biomass burning emissions in urban settings such as Boulder. In both cities, residential biomass burning was prominent in the evening hours. In Boulder, the fraction of VOCs attributed to burning emissions increased during colder nights and was most prevalent in residential areas. In Aspen, domestic burning due to residential heating and cooking was a dominant VOC source at night. When compared to emissions observed in Boulder, biomass burning in Aspen accounted for a higher fraction of nighttime VOCs.
A world avoided: Impacts of changes in anthropogenic emissions on the burden and impacts of air pollutants.

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Abstract:

Emissions from anthropogenic activities are known to have deleterious impacts on human and ecosystem health and as such a significant amount of time, effort and money has been spent developing legislation to minimise their effects. But to what end have these efforts been a success?

Here we use a state of the art coupled chemistry-climate model HadGEM2-ES, with extended tropospheric chemistry, to assess the impacts that changes in emissions from anthropogenic activity have had on the burden and impacts of air pollutants over the last three decades. We use HadGEM2-ES to assess an alternative trajectory in air pollutant emissions to that which we have seen. This alternative trajectory can be consider to reflect a world avoided. In this world avoided, the significant levels of air pollution legislation imposed over the last three decades are simulated to not have come into effect in the contiguous United States and Western Europe. By combining the results of simulations of the world avoided with a base case present day atmosphere our model runs demonstrate that as a result of air pollution legislation, over 500,000 lives a year have been saved owing to reduction in sulfate aerosol and up to 10,000 as a result of improvements in ozone and NO$_2$ pollution. These results highlight the important role of legislation in reducing air pollution related mortality in these areas of the globe and highlight a compelling case for developing regions to follow.
Evaluating the spatial heterogeneity of anthropogenic VOC in São Paulo with other urban worldwide observations: a global comparison of source emission composition.

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Abstract:

Volatile organic compounds (VOCs) are important pollutants present in urban environments which affect air quality producing other secondary pollutants (ozone and SOA). Field observations of VOCs can provide relevant information for a better prediction of their products, likewise for building up emission inventories, which strongly depend on an accurate knowledge of their primary emissions. Several studies developed in northern mid-latitude megacities estimated the urban emission ratios (ER) of several VOCs relative to CO and acetylene from field observations. These calculated ER were successfully used as constraints to evaluate emission inventories, and to assess the spatial variability by comparison to other cities worldwide. In this work we used detailed databases of speciated VOCs collected in contrasted urban environments worldwide. VOCs measurements have been collected during intensive field campaigns in Europe (Paris, London, Istanbul), North America (Los Angeles), Asia (Beijing, Beirut, Mecca), and very recently in São Paulo Megacity (MASP-Brazil) and West Africa cities. Data can either encompass source emission measurements and ambient concentrations. In particular, MASP with more than 20 million inhabitants and 7 million vehicles, is among the world's most populous cities; where ethanol is widely used by flex-fuel vehicles. Even though some short campaigns were carried out, little is known about VOCs emission and composition due to limited temporal and spatial measurements. Since 2012, continuous observations were performed in MASP. A first multi-spatial insight of VOCs composition revealed a similar profile compared to those of other megacities, while they present higher mean values suggesting VOCs distribution is not affected by the large use of ethanol. The global comparison were accomplished in order to elucidate agreements and discrepancies in MASP VOCs profiles and, therefore, understanding spatial heterogeneity of VOCs composition. For that, exploratory tools were applied and ER were estimated and
compared with the values derived from other urban sites.
Abstract:

Atmospheric iron (Fe) plays an essential role in the carbon cycle, affecting the Earth's energy balance and human health. Fe catalyzes oxidations of organic carbon species and serves as a limiting nutrient for phytoplankton in about half of the world's oceans. Wind-blown dust is the major source of atmospheric insoluble Fe while urban areas are correlated with relatively high percentages of soluble Fe. The occurrence of elevated levels of soluble Fe near urban and industrial regions suggests a correlation between Fe solubilization and organic combustion products, including polycyclic aromatic hydrocarbons (PAH). Fossil fuel consumption for internal combustion engines produce atmospheric PAH as a major component of automobile exhaust. Under light, PAH transform into oxidized components such as ketones and carboxylic acids. For example, phthalic acid (formed from naphthalene) inhibits Fe oxidation reactions and therefore may contribute to Fe reduction and increased solubility. The wind-blown dust and PAH-containing combustion products undergo long-range atmospheric transport leading to mixing and metal-organic interactions. The current study focuses on how a saturated PAH suspension affects the production of soluble Fe. Reactions of soil-based Fe and saturated solutions of PAH are performed under controlled conditions simulating natural sunlight. Samples are analyzed by ICPMS for soluble Fe before and after solar exposure reactions; soluble Fe is separated from total Fe by filtration and total Fe by acid-assisted microwave digestion. Data indicate an increase in Fe solubility (1.2% to 4.2%) in the presence of PAH, as compared to soil in water alone, and an even greater increase in Fe solubility (4.2% to 8.4%) when exposed to solar radiation. Research is ongoing to determine the dependence of oxidized PAH on kinetic and overall Fe solubility.
1.119 In-use heavy-duty diesel vehicle emission measurements used to investigate the durability of diesel particulate filters.

Early Career Scientist

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Abstract:

Heavy-duty diesel trucks are important sources of oxides of nitrogen and particulate emissions. The recent introduction of selective catalytic reduction systems (SCR) and diesel particulate filters (DPF) can dramatically lower the levels of both of these when properly maintained. The Ports of Los Angeles and Long Beach instituted a forced retirement program requiring all trucks operating to have 2007 U.S. compliant engines (requiring a DPF) by January 1, 2012. This fleet comprises one of the largest concentrations of DPF equipped heavy-duty diesel trucks making it an ideal fleet for studying particulate emission deterioration. The University of Denver has collected in-use fuel-specific emission measurements from heavy-duty trucks since 2008 at the Port of Los Angeles using optical remote sensing equipment. Beginning in 2013 we began using a new emission collection method that allowed for enhanced information of particle emissions at our Port of Los Angeles location and a new site on I-5 near Cottonwood, CA in Northern California.

The On-road Heavy-duty Vehicle Emission Monitoring System (OHMS) employs a 15.25m long tent and a perforated pipe to capture the integrated exhaust emissions of a heavy-duty truck as it drives through. Gaseous emissions collected include carbon monoxide, carbon dioxide, hydrocarbons, nitric oxide and total oxides of nitrogen. Particle mass and number concentration are measured with a Dekati Digital Mass Monitor while black carbon is quantified using a Droplet Measurement Technologies Photoacoustic Soot Meter. In two field campaigns in 2013 and 2015 we have collected more than 5000 fuel-specific emission measurements from the two sites. These data allow us for the first time to the investigation of the performance and durability of diesel engine emission controls. DPF’s initially produced large reductions in particle mass and number emissions, however, the 2015 data shows increases in both raising concerns as to their durability.
1.122 EMISSIONS AND ATMOSPHERIC CHEMISTRY FROM HAVANA’S STATIONARY SOURCES.

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Abstract:

The determination quantitative of air pollutant emission and dispersion in the atmosphere from main stationary sources in Havana and its effects on atmospheric chemistry, is an urgent and necessary study, taking into the account that mentioned city have the most population of country and a number considerable of emission sources. The knowledge of these emissions and dispersion is a useful tool to know the characteristic the atmospheric chemistry in urbanization. The results showed atmospheric emissions rate of SO$_2$ is upper than 25 thousand ton/year while NO$_2$ emission rate is about 7300 ton/year. Furthermore, particulate matter (PM$_{10}$ and PM$_{2.5}$) reach 2300 ton/year, which are known for its potential damage for human health and atmospheric chemistry. The most populated municipalities are very close to the fixed sources emit more pollutants into the atmosphere. The influence of urbanization is reflected in the dispersion of emissions pollutants. Finally, this air emission inventory is a previous stage before of that will be allow future implementation of air quality forecasting for Havana city from any air pollution models outputs.

**Keywords:** Emission, sources stationary, atmospheric chemistry
First results from a field campaign at a high altitude urban metropolitan area in the Central Andes.

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Abstract:

From the beginning of 2016 instrumentation has been set up at two of the largest cities of Bolivia, La Paz and El Alto, which are part of the metropolitan area of La Paz. Two
locations were chosen in order to study the background urban aerosols: the International Airport of El Alto at 4090 m asl, at the edge of the Altiplano plateau, and the ex-zoo of La Paz at 3600 m asl located at the center of the city within a valley. Instruments for characterizing aerosol particle properties, both physical and chemical, have been deployed at both sites. In addition the concentrations of several reactive gases are being monitored at the same time. Besides studying the temporal behavior of all these quantities, one important goal of the campaign is to estimate emission factors at the peculiar conditions of this region (high altitude, elevated intensities of UV radiation and a large fleet of relatively old vehicles running on gas, gasoline and diesel). Studies related to source apportionment have been designed and will be carried out during the planned one-year campaign. The outcomes of this effort, together with the results of a previous, but shorter study, are expected to help local governments to take measurements in order to improve air quality in the region. In addition, this field campaign will contribute to a better understanding of observations performed at the nearby GAW station of Chacaltaya (5240 m asl) and the transport of pollutants into the free troposphere. The results from the first months of continuous measurements at both locations will be presented and discussed under this framework.
1.126 Sources and Heterogeneous Production of Nitrous Acid and Impacts on Air Quality: Overview of Results from Integrated Field, Lab and Modeling Studies in Hong Kong.

Early Career Scientist

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Abstract:

Nitrous acid (HONO) plays an important role in the chemistry of polluted atmosphere, but its sources in different environments are not well understood. Here we give an overview of our recent efforts in investigating the sources and formation of HONO by combining the field measurement, lab experiments and model simulation. Since 2011, a series of field studies on HONO was carried out at several sites (one sub-urban, one coastal background, one mountain-top, one roadside and one tunnel) in Hong Kong, which is situated on the South China coast. The data are examined to elucidate seasonal characteristic, emission factors, heterogeneous production, and its photochemical impacts. The derived HONO emission factors from vehicles showed high variability, and for most of time, higher than the r uniformed value of 0.8% reported in literatures. Larger nocturnal heterogeneous conversion rates of NO$_2$-to-HONO were observed when air masses passing over sea surfaces than land surfaces (~3 times), suggesting that air-sea interactions may be a significant source of atmospheric HONO which has never been considered in previous air quality model studies. For selected episodes, the strengths of the ‘unknown’ daytime source were estimated, and its correlations with different surfaces suggest that aerosols could play a more important role in HONO production than ground when aerosol loading and humidity is high. Furthermore, a series of flow-tube lab experiments found that SO$_2$ aging on mineral particles could significantly enhance the uptake of NO$_2$ and HONO formation. The up-to-date HONO sources were parameterized
into a WRF-Chem model, to better understand its impacts on air quality. The revised model significantly improved the simulations of ozone, and increased ozone concentrations by 6-12% over urban areas in HK-PRD region. Our studies reveal the complexity of HONO sources and chemistry, and demonstrate its importance in transforming NO$_X$, photochemistry and air quality in polluted regions.
Impacts of the decadal urbanization on thermal circulations and ozone production in the Pearl River Delta region, China.

Early Career Scientist

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Abstract:

Thermal circulations induced by urbanization could exert important effects on ozone (O₃) production through regulating the chemical transformations and transport of O₃ and its precursors. Previous studies lack a correct representation of urban vegetation abundance, and thus is difficult to accurately describe the land-atmosphere coupling. In this study, the Weather Research and Forecasting/Chemistry (WRF/Chem) model combined with Moderate Resolution Imaging Spectroradiometer (MODIS) remote sensing are used to investigate the urbanization impacts on thermal circulations and O₃ production in the Pearl River Delta (PRD) region, China. The assimilation of MODIS land-surface parameters (i.e., land-cover type, green vegetation fraction and leaf area index) provides a clear model improvement for near-surface meteorological variables. A typical urban heat island (UHI) is generated in PRD, which in turn modifies local circulation by initiating the UHI circulation and enhancing the sea breeze over the Pearl River Estuary. Overall, the modified urban meteorology cause a detectable decrease of daytime O₃ concentration (−1.3 ppb) and an increase of O₃ (+5.2 ppb) around the nocturnal rush-hours. The suppressed O₃ titration destruction due to NOx dilution into the deeper urban boundary layer (200−400 m) is the main reason for elevated nocturnal O₃ levels. In the daytime, however, the upward transport of O₃ precursors weakens near-surface O₃ photochemical production and conversely enhances upper-level O₃ generation. Furthermore, the surface UHI convergence flow and intensified sea breeze act to effectively trap O₃ at the suburban and coastal regions. This work may help understand the feedbacks between urban meteorology and air quality.

Presenting Author:

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Abstract:

Automotive fuels and light-duty vehicle technologies have evolved substantially over the past few decades. The fundamental factors influencing this evolution, the likely future trajectory of vehicle emissions, and their impact on urban air quality will be discussed. Emissions of volatile organic compounds (VOCs), CO, nitrogen oxides (NOx), and particulate matter (PM) from internal combustion engines reflect complex processes involving interactions between the fuel and engine parameters. Engine hardware and operating conditions, after-treatment catalysts, and fuel composition all affect the amount and composition of vehicle tailpipe emissions. Fuel volatility and vehicle fuel system design affect evaporative emissions.

Vehicle emissions of VOCs, NOx, CO, and PM contribute to photochemical smog formation in urban atmospheres. There have been large reductions in the emissions from new vehicles over the past several decades. Emissions from the on-road fleet are declining reflecting fleet turn-over as new vehicles with modern emission control systems replace older vehicles. Decreased emissions from traffic have been an important factor in the observed improvements in air quality in U.S. and European cities. As an example, from 1980 to 2014 the national average levels of CO, O₃, Pb, NO₂, and SO₂ in the air in U.S. cities decreased by approximately 85%, 33%, 98%, 57%, and 80%, respectively (USEPA, 2016). The global trend of reduced emissions from road traffic and reduced contribution of vehicle emissions to air quality issues will be presented and discussed.
1.130 Energy, Air Quality, and Urbanization.

Presenting Author: Tracey Holloway, University of Wisconsin--Madison, Madison, Wisconsin, USA, taholloway@wisc.edu

Abstract:

Cities around the world are on the front lines in dealing with today's pressing social, economic and environmental issues. Cities strongly affect how humans use energy - how they drive, how they live, how they work, how they consume goods and services. Energy consumption, in turn, strongly affects air emissions of carbon dioxide (CO$_2$), nitrogen oxides (NO + NO$_2$ = NO$_x$), particulate matter (PM) and other compounds of relevance to climate and/or public health. As the global community considers solutions to climate change, cities have come to be viewed as potentially transformative in reducing global carbon emission. These efforts dovetail with related initiatives to address health-damaging air pollution, which often peaks in densely populated urban environments. The role of cities in global change cannot be overstated. Globally, over half of the population lives in cities; in the United States, over 80% of the population. Cities thus account for the majority of global carbon emissions, and represent the most rapidly changing aspect of energy-relevant global infrastructure. This globally transformative role is all the more surprising considering that cities represent just 2% of the Earth’s surface area.

Recent work will be presented on the characterization and trends of urban energy use, emissions, and air quality. Particular emphasis will be given to the potential for satellite data to support decision-making relevant to air quality.
Inverse modeling and satellite data analysis for improving emission inventories.

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Abstract:

High time resolution surface measurements of pollutant concentrations were used as input to an inverse model in order to constrain emission estimates from a range of sources. By using a combination of particle backward trajectories, forward grid simulations and tracer concentrations, an inverse model was used to estimate the diurnal and seasonal variation of anthropogenic emissions as well as biomass burning emission factors for species such as mercury and carbonaceous aerosols. Shifting from in-situ measurements to remote sensing, satellite retrievals were used to estimate trends in emissions for urban areas and industrial point sources. A multiple linear regression model was developed to analyze long time series of individual pixel data from satellite retrievals. This enabled the identification of day of week and seasonal variability as well as the impact of the economic cycle and the implementation of control policies on year to year variability. Examples will be shown using OMI NO2 data for areas around the world.
Chemical characterization and source apportioning of Particulate Matter (PM10 and PM2.5) during winter fog season (2015-2016) in the three megacities of Pakistan.

Early Career Scientist

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Abstract:  

The Asian region over the last decade have undergone rapid economic development along with urbanization, motorization and extensive energy use. As a result air pollution emerged as a serious threat to the environment and population health in the region (Gurjar et al. 2008)(Hopke et al. 2008). According to different studies 865,000 premature deaths caused every year by air pollution and about 60% of these deaths found to be occurred in Asia (WorldHealth2007). Pakistan in the region is among the countries effected more by air pollution. The hazardous impacts of air pollution becomes worse during the winters due to dense persistent fog especially in the months of December January and February (DJF). It has been reported that the average concentration of both PM10 and PM2.5 increased about two times during intense fog days. Many parts of Pakistan experienced severe fog and haze during the months of DJF that lead to significant social and economic problems specially the Disruptions of road and air traffic. A three months long land campaign was undertaken in three megacities of Pakistan namely Lahore, Sheikhopora and Faisalabad. The selection of these cities were based on the fact that these cities are always more vulnerable to winter Fog, and every year lots of accidents and economic losses reported from these regions. The main objective of this study was to evaluate the concentrations and chemical characterization of particulate matter during winter Fog season. Aerosols were collected at different locations by using High/low volume samplers. For elemental composition ED-XRF was used. In next step AAS was used to quantify different metals detected by ED-XRF. Positive Matrix Factorization (PMF) model is an effective source apportionment receptor model that was used to find out the possible sources of PM10 and PM2.5. For better analysis and validate sources, back trajectories were also calculated.
1.139 Hemispheric and regional source-receptor relationships for air pollution in East Asia.

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Abstract:

Within the UN Convention of Long-range transported air pollution (LRTAP), the EMEP MSC-W chemical transport model has been used for source-receptor calculations (SR) for several decades (published at the http://emep.int website). SR relationships quantify the contributions of emissions from one country to air pollution and depositions in another. SR relationships are presented annually for Europe and western parts of Asia. Since recently, SR relationships have been calculated on regional scales, down to large cities and agglomerations, also for regions in East Asia.

This presentation will review results obtained with the EMEP MSC-W model within the Task Force on Hemispheric Transport of Air Pollution (organized under the LRTAP convention) and for the EU project PANDA for East Asia. Relevant questions to be addressed are 1) how much of the regional air pollution in East Asia is due to indigenous emission sources and how much is due to long-range transport (e.g. from Europe or North America) and 2) how much of the air pollution in large cities (e.g. the megacities of Beijing and Shanghai in China) is indigenous and how much is imported?

These questions are of high relevance for air quality policy makers, both as a basis for decisions on local and regional emission reduction measures and for the assessment of compliance with air quality standards. On episodical time scales (a few weeks or less) source-receptor relationships depend not only on the agglomeration-specific distribution of emissions but also on meteorological conditions that can vary strongly from day to day. Thus, SR relationships need to be re-assessed regularly (e.g. weekly), as an important supplement to air pollution forecasts, or as reanalyses of specific air pollution episodes in the past.

Early Career Scientist

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Abstract:

Photochemistry experiments were performed with the potential aerosol mass (PAM) reactor at Colorado State University in the summer of 2015. The goal was to investigate primary emissions and secondary production of organic aerosol (OA) from a representative non-road diesel engine under varying engine load and fuel combinations. SOA production dominated POA emissions by an order of magnitude after 1.5 days of simulated atmospheric aging, idle loads produced the highest POA and SOA levels and biodiesel results were identical to those from diesel. In this work, we used two state-of-the-science frameworks, namely the volatility basis set (VBS) developed by Donahue and coworkers and the statistical oxidation model (SOM) developed by Cappa and coworkers to simulate the production and composition of SOA in those experiments. Leveraging recent laboratory-based inputs, both frameworks accounted for a semi-volatile and reactive POA, SOA production from volatile organic compounds (VOCs) and intermediate-volatility organic compounds (IVOCs), NO\textsubscript{X}- dependent multigenerational gas-phase chemistry and dynamic gas/particle partitioning. Both model frameworks demonstrated that for model predictions of SOA mass and composition to agree with measurements across all engine load-fuel combinations, it was (a) quintessential to account for SOA formation from IVOCs (IVOCs were found to account for about three-quarters of the model-predicted SOA) and (b) necessary to dynamically model the gas/particle partitioning using a mass accommodation coefficient of approximately 0.1. Model predictions of the gas-phase organic compounds (resolved in carbon and oxygen space) from the SOM compared favorably to gas-phase measurements made using a Chemical Ionization Mass Spectrometer (CIMS). Qualitatively, this finding substantiates the semi-explicit chemistry captured by the SOM and the measurements made by the CIMS. Work is under way to examine the sensitivity of the findings to the use of alternate emissions profiles and treatment of vapor wall-losses.
1.146 Use of concurrent top down approaches to assess emissions inventories for the central Mexico cities belt.

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Abstract:

The Mexico national emissions inventory 2008 (INEM-2008), the latest available for nationwide air quality modeling, was evaluated for the central Mexico city belt (CMCB). Two, complementary top-down approaches were used to assess the high resolution emissions inventory model fed to WRF-Chem.

First, one month observations between February 6 to March 8 2011, made in three peri-urban monitoring sites: Tenango del Aire, Amecameca and Ozumba, municipalities in the State of Mexico, southeast of Mexico City Metropolitan Area (MCMA), at the Tenango del Aire Mountain Pass, were used to evaluate WRF-Chem. Two exclusive kinds of episodes, when the sites are down or up wind from MCMA used.
Prior to any air quality modeling, several land use data-bases were assessed to find the one that best reproduces temperature and wind fields for the region using WRF. We found that the USCS NALCMS2005 data base gave the best results. Then, slow-reacting CO output from WRF-Chem was used to test several grid-nesting choices. We settled for an inner domain made of a 3x3 km$^2$ grid with 89 x 89 containing the central region of Mexico. The external domain was a 90 x 90 km$^2$ grid covering most of the country. At this stage CO is severely underestimated, for more than a factor of 10 when under local emissions influence and by a factor of 5 under regional influence.

The second top-down approach is comparison with satellite observations. February to June 2011 output from WRF-Chem, using the above model configuration, is compared against the IASI CO sensor on board of Metop A. Preliminary comparisons indicate CO columns from the model above MCMA are 0.7 lower than observations, but overall in the region they are underestimated by a factor of 3. Work is underway to improve on these comparisons and to complement results from both methods.
Response of winter fine particulate matter concentrations to emission and meteorology changes in North China.

Early Career Scientist

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Abstract:

The winter haze is a growing problem in North China, but the causes have not been well understood. The chemistry version of the Weather Research and Forecasting model (WRF-Chem) was applied in North China to examine how the PM$_{2.5}$ concentrations change in response to changes in emissions (sulfur dioxide (SO$_2$), black carbon (BC), organic carbon (OC), ammonia (NH$_3$), and nitrogen oxides (NO$_x$)), as well as meteorology (temperature, relative humidity (RH), and wind speeds) changes in winter. The sensitivity results indicate SO$_2$, OC and NH$_3$ emissions should be controlled preferentially to control winter haze. Based on the major sources of SO$_2$, OC and NH$_3$ emissions, natural gas should be promoted to take the place of coal and biofuel, and some animal feeding and animal housing strategies should be taken to control NH$_3$ emissions. The perturbations in temperature, RH and wind speed do not show significant impacts on numbers of haze days, suggesting that the haze occurrences are more likely to be associated with atmospheric circulation.
Analyses of the main sources of PM2.5 using bottom up and top down data in two metropolitan cities in Mexico: Mexico City Metropolitan Area and Toluca Metropolitan Area.

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Abstract:

Although important improvements have been achieved in Mexico City Metropolitan Area (MCMA) air quality over the last three decades, ozone and PM2.5 are still above the national air quality standards and WHO recommendations. Air quality monitoring in other cities has been expanded in recent years; these data show that smaller cities and metropolitan areas, where air pollution was not perceived as a problem, are facing high levels of pollutants, with PM2.5 levels even two to three times higher than those registered in MCMA since 2003. Although PM2.5 health impacts are well evidenced, existing air pollution contingency programs in Mexican cities do not include PM2.5 yet, mainly because identification of effective control measures are still lacking. In this work, an analysis of the main PM2.5 emissions sources is presented using existing bottom-up inventories and when available, top-down emissions studies for two Mexican metropolitan areas: the Mexico City Metropolitan Area (MCMA) and Toluca Metropolitan Area (TMA). TMA is metropolitan area with 2.1 million habitants (2,203 km$^2$) comprises 14 counties including the capital city of the State of Mexico and it is connected economically and physically with MCMA. Annual average levels for the TMA and MCMA were 42 µg/m$^3$ (in 2012) and 27 µg/m$^3$ (in 2014). PM2.5 levels in MCMA (18 million people/ 7,866 km$^2$) have been maintained over the last decade in contrast to the increment in population and vehicular fleet. MCMA bottom up inventories indicate residential combustion and soil resuspension as the main PM2.5 sources (53%), top-down emissions identify other sources contribution. TMA bottom-up inventory indicates residential wood combustion (cooking) as the main PM2.5 emissions sources. In this work, a proposal of a top-down emission analyses integrated by a group of institutions including a field campaign and remote sensing measurements will be presented for TMA.
**1.149 Implementation of a high-resolution WRF-Chem CO tracer model in Buenos Aires, Argentina.**

Early Career Scientist

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Abstract:

This study presents the implementation of a state-of-the-art atmospheric chemistry model over the Buenos Aires Metropolitan Area of Buenos Aires (MABA). The MABA is a highly populated area with an estimated of more than three million of vehicles circulating in approximately four thousand square kilometers. Even though the MABA has a good vent due to the flat surroundings and regional atmospheric circulation, it is important to monitor and study the evolution of pollutant concentrations given the intensity of the local sources and the size of the population potentially exposed to harmful concentrations of contaminants. Few studies dealt with the high resolution three dimensional atmospheric modelling of pollutants in MABA region. In this work we implemented the WRF-Chem model over MABA region using a high resolution area source emission inventory of carbon monoxide (CO) (Venegas et al. 2011). The evaluation of the model performance in reproducing CO concentrations at several observations stations within the city, under different large scale environmental conditions is presented. The emission inventory for the MABA includes mobile sources (road traffic and airplanes) and fixed sources (residential, commercial and small industries activities). The test cases chosen were: 10-14 November of 2009 and 2-7 June 2010 where we have available and continuous measurements at least two different locations within MABA. In these first experiments we model carbon monoxide concentrations considering it as a passive tracer. The configuration of the model includes 2 nested way with an inner domain of 1 kilometer in which we test the sensibility of the modeled concentrations to three different boundary layer parameterizations (YSU, MYJ and MYNN) representing the most common schemes used in WRF-Chem. Results showing the evolution of CO concentrations as well as the comparison with the observations will be presented at the conference.
Abstract:

The kinetics of oxidation of carbon-monoxide on the nano-catalyst surface in different conditions has been investigated. The laboratory experiments have done in quartz reactor is installed cycle system (d=10 mm, h=1 m) where is controlled speed of air stream, the temperature of surface of catalyze and change of concentration of carbon monoxide and carbon dioxide in air mixture. Experiments carried out by preliminarily prepared model mixtures of CO with air. Experimental plants allow to change temperature on surface of nano-catalyzer, flow rate of mixture air-CO and concentrations of gases CO and CO2 in the closed system. The investigation has been conducted in flow rate 38 l/min and temperature range of $T=70-350\,^\circ\mathrm{C}$, the dependence of conversion degree of carbon monoxide to carbon dioxide on experimental parameters has been controlled. It is determined that rate of conversion process increases by 1.30-1.32 times as temperature increases on the surface of catalyzer. The conversion of carbon-monoxide to carbon dioxide increase by decreasing of flow rates. The carbon monoxide decreased up to % under optimal condition, that has a great importance to prevention of air pollution by transport sector.

The environmental impact assessment of transport sector to the air basin of Azerbaijan has been assessed and decreasing of impact by application of nano-catalyst purification technology are estimated.
Pollution over Megacity Regions from the Tropospheric Emission Spectrometer (TES).

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Abstract:
The world’s megacities, defined as urban areas with over 10 million people, are growing rapidly in population and increasing in number, as the migration from rural to urban areas continues. This rapid growth brings economic opportunities but also exacts costs, such as traffic congestion, inadequate sanitation and poor air quality. Monitoring air quality has become a priority for many regional governments, as they seek to understand the sources and distribution of the species contributing to the local pollution. Hyperspectral infrared instruments orbiting the Earth can measure many of these species simultaneously, and as they measure averages over their footprints, they are less sensitive to proximity to strong point sources than in situ measurements, and thus provide a more regional perspective.
The JPL TES team has selected a number of megacities as Special Observation targets. These observations, or transects, are sets of 20 closely spaced (12 km apart) TES observations carried out every sixteen days. We will present the TES ozone ($O_3$), peroxycetyl nitrate (PAN), ammonia ($NH_3$), formic acid (HCOOH) and methanol (CH$_3$OH) data collected over Mexico City and Lagos (Nigeria) from 2013 through 2015. If time permits we will also present data over other cities in or near biomass burning regions, such as Delhi. We will combine the TES data with MODIS AOD and AIRS CO, as well as back trajectories, to determine to what degree the seasonality and spatial gradients in the TES measurements are driven by local emissions, biomass burning and regional circulation patterns. Some of the transects demonstrate very nicely the synergy obtained from simultaneous measurements of multiple trace species. We will also discuss how the spatial variability along the transects is related to topography and land use.
1.155 Spatial and temporal trend of Persistent Organic Pollutants in airborne particle matter in Metropolitan Area of Mexico City.

Early Career Scientist

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Abstract:

Persistent Organic Pollutants (POPs) are a group of halogenated synthetic compounds widely used in common applications as pesticides, dielectric fluids and flame retardants. A large number of compounds are prohibited or restricted by the Stockholm Convention due to their high toxicity, risk to human health and environmental pollution. A simple, fast and green methodology was developed to determine POPs in airborne particle matter using a micro scale cell and ultrasonic assisted extraction. POPs were obtained in an online extraction-filtration-evaporation system and analyzed by gas chromatography – mass spectrometry/negative chemical ionization. Application of this methodology allowed us to study temporal and spatial POPs trend in five urban sites in Metropolitan Area of Mexico City (MAMC) by collecting particles ≤ 2.5 µm (PM$_{2.5}$) in 2013. BDE-99 (pentabromodiphenyl ether), endosulfan and endosulfan isomers were found in all sites and seasons. Endrin aldehyde, dieldrin, DDE and other polybrominated diphenyl ethers (PBDEs) were also found but they were not uniformly distributed. Temporary homogeneous distribution of organochlorine pesticides sum was observed (Kruskal-Wallis, KW, $p = 0.8$). Polybrominated diphenyl ethers sum showed lower concentration in rainy season compared with warm and cold dry seasons (KW, $p \leq 0.02$). Northeast and Southeast showed higher pesticides sum concentrations than Center, Northwest and Southwest of MAMC (KW, $p \leq 0.01$). PBDEs sum was higher in Northeast respect to the rest of MAMC (KW, $p \leq 0.004$). Results showed evidence of POPs contamination in MAMC and for our knowledge this is the first time that POPs were determine in airborne particle matter in urban sites from Mexico.
1.156 Air pollution in Mexico City from remote sensing measurements.

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Abstract:

The Mexico City Metropolitan Area (MCMA), with over 21 million inhabitants and around 7 million motor vehicles filling the streets, faces air pollution problems which are worth investigating not only at its surface. A broad remote sensing infrastructure has been set up to increase the vertical information of key pollutants such as CO, NO₂, O₃, CO₂, CH₄ and NH₃ from ground-based solar absorption measurements in the infrared as well as in the UV/visible regions. In this contribution we will discuss the advantage of using column measurements in synergy with satellite observations and surface concentrations to better understand the air quality in a complex setting such as the MCMA. Particular emphasis will be placed in the results for carbon monoxide (CO). Its annual variability in central Mexico is related to biomass burning activities mostly in the dry season, but distinct weekly pattern and diurnal cycles in the urban environment are
dominated by the emissions from the transportation sector. A detailed study of the mean vertical and spatial distribution of CO enables the tracing of sources and helps to understand the transport mechanisms. Total vertical columns and profiles are measured outside the city at a high-altitude site (Altzomoni, 3985 masl) and within the city (UNAM campus: 2260 masl) only 60 km apart. The evolution of the mixing layer height, which is critical for understanding ground level concentrations, is investigated by means of a low-cost LIDAR and a reconstructed product from both surface and column integrated measurements. A satellite carbon monoxide product is used to obtain the spatial distribution of CO around the MCMA. The trends and variability of this pollutant will be shown and the potential of using a rich set of observations like these to reconstruct the emissions will be discussed.
1.157 An investigation of air pollution levels along selected roads in Nairobi, Kenya.

Early Career Scientist

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Abstract:

This paper presents a statistical analysis of air quality monitoring at three major roads and Industrial Area in Nairobi. The study was carried out using various gas analyzers and samplers mounted in a Mobile Air Monitoring Laboratory (MAML) van. The report realized extremely high values of black carbon which went beyond the upper limit of the instruments (50,000 ng/m$^3$) during the day in Landhies road. Nakumatt Junction recorded extreme values of Black carbon (14,008 ng/m$^3$) in the evening peaks while Pangani Roundabout, the diurnal mean value was extreme (14,446.5 ng/m$^3$). None of the four sites exceeded the WHO 24 hour limit for both PM$_{10}$ (50 μg/m$^3$) and PM$_{2.5}$ (25 μg/m$^3$). The 24 hour mean of PM10 in the three sites also did not exceed the ambient air quality tolerance Kenyan limit of 100 μg/Nm$^3$ and 150 μg/Nm$^3$ in Industrial Area. Oxide of nitrogen showed two pronounced peaks, one in the morning and the other in the evening. The diurnal mean of SO$_2$ over the four sites was generally low with the highest amount of 1.08 ppb recorded at Pangani Roundabout. It is recommended that efforts be made to maintain or even reduce further the present pollution levels, meaning concerted efforts need to be made to find a sustainable balance between industry, human health and environmental protection.
Spatial distribution of gaseous pollutants (NOx, SO2, NH3, HNO3 and O3) in Abidjan, Côte d'Ivoire.

Early Career Scientist

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Abstract:

This work is part of the DACCIWA FP7 program (Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa) in the framework of the work package 2 « Pollution and health ». This study aims to characterize urban pollution levels through the measurement of NO2, SO2, NH3, HNO3 and O3 in Abidjan and Cotonou, the economic capitals of Côte d'Ivoire and Benin. Gases measurements are performed using passive samplers exposed in duplicate for two weeks periods. Our study allowed since December 2014 a monitoring for a two years period (2014-2016) at three sites in Abidjan and one in Cotonou; all representative of major sources of pollution (traffic, domestic fires, waste burning). In addition, we performed an intensive measurement campaign in Abidjan from December to February 2016 during the dry season. Sixteen sites in the district of Abidjan were selected to be representative of various anthropogenic and natural sources of the city. Our results document the interannual and seasonal variability of gaseous pollution at the monthly scale. Results from the intensive campaign show that the gases concentrations are strongly linked to pollution sources nearby and show a high spatial variability of gaseous pollutants concentrations on different sites of Abidjan. However three gases present higher levels of concentrations at all the sites: NH3 (53%), NO2 (21%) and O3 (21%). NH3 concentrations vary between 102,1±12,1ppb measured at the domestic fire site and 13,6±3,5ppb measured at the suburban site. NO2 mean concentration vary from 19,6±1,7ppb to 5,4±0,93ppb. We measured the two most important O3 concentration on the two coastal sites (19,4±2,3 and 19,1±3,8). The average concentrations of SO2 never exceeding 1,3ppb at all the sites. All these results will be combined with meteorological parameters to provide a first mapping of gaseous pollutants at the scale of these two West African capital. These measurements constitute an original database because no air
quality network.
Chemical composition and source apportionment of PM2.5 in Beijing based on daily samples collected in 2012-2014.

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Abstract:
Beijing, the capital of China, has been experiencing serious air pollution over the last decade. A variety of governmental policies have been implemented to improve the air quality, however, the concentration of PM$_{2.5}$ remains high. Understanding the sources of PM$_{2.5}$ is of essential importance for air quality management. From 2012 to 2014, daily PM$_{2.5}$ samples were collected in an urban site for 24 months in Beijing, China. The major chemical components of PM$_{2.5}$ analyzed include water-soluble ions, organic carbon (OC), element carbon (EC), and trace elements. Seven main components were identified, including sulfate, nitrate, ammonium, minerals, trace element oxidizes (TEOs), organic matters (OM) and EC, and the average concentration were 11.7 (0.1-39.7), 13.2 (0.2-95.8), 6.7 (0.1-39.8), 11.9 (0.4-227.0), 3.6 (0.1-26.7), 23.5 (2.4-106.0), 1.7 (0.1-8.9) μg m$^{-3}$, respectively. Distinct seasonal variations were clearly seen. The positive matrix factorization (PMF) model was applied for the source apportionment of PM$_{2.5}$. Six PM$_{2.5}$ source categories were identified. The major contributors of PM$_{2.5}$ were secondary inorganic aerosol, followed by coal combustion, biomass burning, vehicle emissions, minerals, smelting industry. Seasonal variations of the contribution of each sources were shown. Minerals contributed more in spring than in other seasons in 2012, but not in spring in 2014, which may mainly due to the changes in wind speed and direction. The contributions from coal combustion and biomass burning were higher in heating seasons than in non-heating seasons, The major reasons for this seasonal variation could be straw burnnings for cropland cleaning as well as wood burnnings and coal combustion for heating. The contribution from secondary inorganic aerosol was the largest and did not show that obvious seasonal variations compared to other sources. This may be caused by the higher emissions of NO$_2$ and SO$_2$ in winter and the strong photochemical reaction in summer.
An estimation of NOx emissions from OMI-observed NO2 columns over East Asia.

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Abstract:

For the high-resolved (i.e., 30km x 30km) top-down NO\textsubscript{x} emissions, an algorithm was developed based on the mass balance equation. Two main parameters were incorporated in the algorithm. For the first, atmospheric NO\textsubscript{x} molecules transported from/to the adjacent cells for the considering the non-local sources were sophisticatedly calculated. For the second, effective NO\textsubscript{x} lifetime for the nonlinearity between NO\textsubscript{2} columns and NO\textsubscript{x} emissions was estimated. In our analysis, the NO\textsubscript{x} transports from/to the neighborhood cells had significant impacts on the effective NO\textsubscript{x} lifetime in both cold and warm seasons. Also, in our sensitivity test, we showed that the errors in the top-down NO\textsubscript{x} estimations can be reduced by filtering the data whose NO\textsubscript{x} lifetimes are smaller than 5 hours. The relative errors caused by the uncertain issues of NO\textsubscript{x} lifetimes with interpolation of satellite data were ~13% and ~5% in January and July, 2014. Using the algorithm, the top-down NO\textsubscript{x} emissions were estimated to be 1.04 and 1.18 Tg N /month over our entire domain for January and July, respectively. The values corresponded to decreases by ~15% and ~2%, compared with the bottom-up NO\textsubscript{x} emissions in January and July, respectively. We also compared the CMAQ-estimated NO\textsubscript{2} columns with OMI-retrieved NO\textsubscript{2} columns to evaluate the bottom-up NO\textsubscript{x} emission (i.e., MICS-Asia III) and investigate how much the top-down NO\textsubscript{x} emissions estimated from our algorithm were improved.
Urban areas of central and southern Chile exceed particulate matter air quality thresholds.

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Abstract:
An analysis of air quality is presented concerning concentrations of sub-10 µm and sub-2.5 µm particulate matter (PM$_{10}$ and PM$_{2.5}$, respectively) recorded at 23 automated public monitoring stations located in 15 cities in central and southern regions of Chile. In each city, the spatial and temporal distributions of PM$_{10}$ and PM$_{2.5}$ concentrations were recorded. Air quality was evaluated via comparison of the annual average concentrations of PM$_{10}$ and PM$_{2.5}$ with the guidelines of the World Health Organization (WHO) and national standards. The results showed that the limits established in the WHO guidelines and the national standards were systematically exceeded at all of the study sites. The greatest concentrations, for both PM$_{10}$ and PM$_{2.5}$, were observed during the fall and winter months (April to September). During the winter months, the burning of firewood for heating produces emissions that are an important source of particulate matter, primarily at night when the lowest temperatures occur and atmospheric conditions are generally unfavorable for dispersion, resulting in the accumulation of pollution above cities. This evaluation of air quality showed that in cities in southern Chile, the population is exposed to concentrations of particulate matter that can have negative impacts on health.

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Clean Air and Urban Landscapes: Towards a Clean Air Plan for Western Sydney.

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Abstract:

The negative health impacts of airborne particulates on urban populations are now well established. Whilst the air quality in Australian cities is generally very good compared to many other parts of the world, Sydney experiences occasional poor air quality events that expose the population to heightened health risks. The population within the Sydney basin is predicted to grow by ~ 20% in the next decade, increasing both the local sources of pollution and the population exposed.

The Clean Air and Urban Landscapes hub has formulated a project to address this issue called the Western Air-Shed Particulate Study for Sydney (WASPSS). The project will provide the tools and evidence to develop a Clean Air Plan for Western Sydney.

Research activities will be focussed around three main research aims, each with their own subprojects:
1. Extend air quality measurement/monitoring capacity to support improved characterisation of air quality in Western Sydney.
2. Implement and validate state-of-the-science regional air quality modelling to assess major sources contributing to air pollution in Western Sydney and assess benefits arising from a range of possible mitigation measures.
3. Evaluate existing methods applied internationally and locally for estimating human exposure to airborne pollutants, and estimate air pollution related exposures and health effects for Western Sydney.

The project will focus on filling measurement gaps to test a new model ensemble as well as estimating human exposure. The project will enable policy makers to estimate the benefits of different mitigation strategies to the future air quality in Western Sydney.
1.169 Are Selective Catalytic Reduction Systems on Diesel Engines an Atmospheric Source of Isocyanic Acid?

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Abstract:

Diesel engines account for half of the nitrogen oxide (NO\textsubscript{x}) emissions from combustion sources in the United States. Strict emissions standards have mandated the use of after treatment devices such as the Selective Catalytic Reduction (SCR) system. In an SCR system, urea is injected into the hot exhaust to yield ammonia, which reduces NO\textsubscript{x} over a catalyst surface to form N\textsubscript{2}. However, SCR chemistry is known to produce isocyanic acid (HNCO) as an intermediate product and SCRs have been implicated as an atmospheric source of HNCO. HNCO is a highly toxic gas linked to adverse health outcomes. In this work, we measure HNCO emissions from a representative diesel engine and, by leveraging earlier data, use a three-dimensional air quality model to simulate the ambient concentrations of HNCO in a polluted urban environment. Experiments were performed on a diesel engine that was configured to meet three generations of EPA emissions standards over the past 10 years. Engine tests were conducted at three different engine loads, with two different fuels (diesel and biodiesel) and four urea injection rates. HNCO was measured using an acetate reagent based chemical ionization mass spectrometer. The engine was found to emit primary HNCO, although the emission factors were an order of magnitude higher than those found earlier; we suspect that steady state engine loads produce more HNCO. In contrast to earlier findings, we do not find any evidence that the after treatment devices produced or enhanced HNCO in the exhaust. The use of biodiesel was found to slightly reduce HNCO emissions. At present, work is underway to model the emissions, chemistry and deposition of HNCO in the UCD/CIT air quality model. The air quality model will be used to predict ground-level concentrations of and comment on the possible human exposure to HNCO in the Los Angeles airshed.
Development of a reactive plume model and its applications.

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Abstract:

A reactive plume model (RPM) was developed to comprehensively consider power-plant plume photochemistry with 255 condensed photochemical reactions. The RPM can simulate two main components of power-plant plumes; turbulent dispersion of plumes and compositional changes of plumes via photochemical reactions. In order to evaluate the performance of the RPM developed in the present study, two sets of observational data obtained from the TexAQS II 2006 (Texas Air Quality Study II 2006) campaign were compared with RPM-simulated data. Comparison shows that the RPM produces relatively accurate concentrations for major primary and secondary in-plume species such as NO$_2$, SO$_2$, ozone, and H$_2$SO$_4$. Statistical analyses show good correlation, with correlation coefficients (R) ranging from 0.61 to 0.92, and good agreement with the Index of Agreement (IOA) ranging from 0.70 to 0.95. Following evaluation of the performance of the RPM, a demonstration was also carried out to show the applicability of the RPM. The RPM can calculate NO$_x$ photochemical lifetimes inside the two plumes (Monticello and Welsh power plants). Further applicability and possible uses of the RPM are also discussed together with some limitations of the current version of the RPM.
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Abstract:
In Southern Chile (south of 34°S), fast urbanization, climate conditions and market fuel prices promote generalized residential wood burning for heating and cooking. However, most households have poor thermal insulation and use inefficient stoves so wood smoke is a widespread problem, with most southern Chilean cities having annual ambient PM2.5 concentrations exceeding 30 ug/m³. Nonetheless, little is known regarding indoor PM2.5 concentrations in that region of Chile. This piece of information is essential to estimate total population exposure to PM2.5.

In this work we consider Temuco (38°44’ S, 72°36’ W, population: 350,000), one of most polluted cities in Chile. We report here the results of indoor PM2.5 measurements carried out in 64 households in Temuco during winter 2014; this is the first time that a comprehensive indoor air quality campaign is conducted in southern Chile. A pair of TAS samplers (Airmetrics, Eugene, OR, USA, 5 L/min) was placed inside each household studied and an additional pair of samplers (Partisol 2000i Thermo Scientific, USA, 16.67 L/min) was placed at a fixed outdoor location for measuring trace elements and elemental and organic carbon in Teflon and quartz filters, respectively. Indoor temperature, relative humidity and CO2 sensors were also placed in each household. In some households, a Dustrack monitor was used to measure indoor PM2.5 continuously. On average, indoor PM2.5 was 90% of outdoor value, which means outdoor PM2.5 easily infiltrates into the indoor environment. We have found that indoor PM2.5 concentrations closely followed the outdoor ones, usually with a delay of 1-2 h, except for a morning peak generated by indoor sources. We present an analysis of PM2.5 infiltration within households and compared them with results from the literature.

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Abstract:

Black carbon (BC) is the second most important species for climate forcing after carbon dioxide. BC deposited in the Arctic can lead to accelerated ice melting. We report the transport and deposition on Arctic ice from wildfire-emitted BC in Northern Eurasia during a 12-year period of 2002 to 2013. We first developed daily BC emissions from wildfires in Northern Eurasia at a 500 m × 500 m resolution during this period. BC emissions were estimated based on the MODIS land cover maps and detected burned areas, the Forest Inventory Survey of the Russian Federation, and emission factors of BC for different types of wildfires. Annual BC emissions from wildfires varied considerably with an average of 0.82±0.50 Tg. BC emissions were dominated by forest fires which accounted for about two-thirds of the emissions, followed by grassland fires (15%). More than 90% of the BC emissions from forest fires occurred in Russia. Central and Western Asia was the major region for grassland fire emissions. Overall, Russia contributed 83% of the total BC emissions from wildfires in Northern Eurasia. The transport and deposition of BC on Arctic ice was simulated daily during the 12 years using the French LMDz-OR-INCA global chemistry-aerosol-climate model. The model has 39 hybrid vertical levels and a horizontal resolution of 1.29° longitude × 0.94° latitude. The model was run using 6-hourly ERA Interim Re-analysis data. The results show that about 7.9% of emitted BC from wildfires in Northern Eurasia was deposited on the Arctic ice, accounting for 45-78% of the BC deposited from all sources. About 20% of the BC deposition occurred in springtime, which is the most effective period for acceleration of melting of ice. The simulated BC concentrations were consistent with observations at the Arctic monitoring stations of Albert, Barrow, Nord, Zeppelin, and Tiksi.
2.004 Ozone Enhancement and Attribution to Wildfires: A Study in the Colorado Front Range.

Early Career Scientist

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Abstract:

Ozone plays an important role on the oxidation capacity and radiative forcing of the atmosphere and at ground-level has negative impacts on human health and ecosystem processes. The Colorado Front Range (CFR) is a region of intricate interactions between multiple pollutant sources and complex meteorological conditions which can result in the accumulation of ozone at ground level. Biomass burning and wildfires have been known to emit a suite of particulate matter and gaseous compounds into the atmosphere which can affect the photochemical processes of ozone. The CFR experiences frequent fires and is impacted by transport of fire related pollutants during the May-September fire season. The extent to which biomass burning pollutants impact air quality depends on many contributing factors, such as, fire size and age, type of material burned, proportions of emitted gas, particulate matter, meteorological conditions, and available NOx and VOC’s. As the climate in the Western United States warms and dries, the conditions and regime of fires and dominant vegetation will be altered. This study demonstrates the importance of understanding the influence of biomass burning on surface ozone mixing ratios in a rapidly changing climate. High ozone events in the CFR associated with fires are analyzed to develop understanding of the influence and variability of ozone and wildfire relationships. A variety of models, satellite imagery, and co-located in-situ measurements are used to characterize the surface ozone conditions in the CFR during wildfire season. These additional measurements and models allow for in depth investigation and understanding of the air parcel origin, chemistry, and movement. This study provides analysis of the frequency and conditions conducive to enhanced ozone episodes which can be confirmed to be transported within and affected by the chemicals and conditions of fires and smoke plumes.
Crop residue burning is a common practice for reducing the volume of combustible materials resulting from agricultural activities in Mexico and Central America. A characterization of the spatial and temporal distribution of agricultural burning is required to assess its environmental impacts and to support fire management. The spatial and temporal variation of fires were investigated using combined multiple satellite observations, focusing on agricultural burning. The burned area approach combines 500 m Moderate-Resolution Imaging Spectroradiometer (MODIS) burned area product (MCD45) with a MODIS Land Cover product (MCD12Q1) and a map of North American land cover produced by the North American Land Change Monitoring System (NALCAMS), from 2001 to 2014. Monthly fire counts were analyzed for eight countries, and three case studies within the Mexican territory were selected for further analysis. The smoke emission from crop burning were analyzed using the Ichoku and Kaufman method.

The annual total number of fire counts in Mexico ranged from 30,489 to 141,061 during the 14 years. Agricultural fires on average accounted for 37% of total detected burns. In Central America, MODIS detected a total annual maximum of 26,012 fires in 2003 and a minimum of 7128 in 2012. Agricultural burning accounted for an average of 43% of all burning in Central America. While the spatial distribution of fires in the eight countries was similar among the 14 years, notable interannual variability was observed in the total number of fire events.

An analysis of the spatio-temporal distribution within Mexico indicates that agricultural fire activity exhibits different seasonality in the north, center and south. In the north of Mexico, monthly agricultural burning counts showed two peaks, one during April to May, and the other in November to December. In the center and the south of Mexico and in Central America, only the former peak is observed.
Abstract:

Isoprene plays a critical role in air quality and climate. Photosynthesis and formaldehyde (HCHO) both hold quantitative information on isoprene emission at large spatiotemporal scales, but neither is a perfect proxy due to their different responses to meteorology. Here, we apply multiple observational data sets: satellite-based solar-induced chlorophyll fluorescence (SIF), flux-derived gross primary productivity (GPP), in-situ measured isoprene emission, and four satellite-based tropospheric HCHO column products to examine the impact of water deficit on the three interlinked processes during the 2012 severe central U.S. drought. At the Missouri Ozarks site (38.74°N, 92.20°W), the 2012 July-to-August SIF, GPP and isoprene emission show substantial reductions of 26%, 80% and 60%, respectively, relative to climatological year values. For the four satellite HCHO products, no concomitant reduction was observed. Possible reasons for the decoupling of isoprene emission and HCHO columns in drought conditions may include: high temperatures accelerate the HCHO production despite less isoprene precursor; lack of precipitation reduces scavenging of reactive oxidants and nitrogen oxides thus facilitating HCHO formation; large retrieval uncertainties result in insensitivity of vertical column concentration to surface perturbation. Our results suggest that satellite HCHO column is not a good proxy for surface isoprene emission under severe drought conditions.
2.009 Long-term measurement of isoprene in a South East Asian tropical rainforest. Initial results and conclusions .

Early Career Scientist

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Abstract:

Isoprene, dominated by emissions from tropical forests, contributes the largest source of reactive carbon to the atmosphere, resulting in changes in surface ozone and secondary organic aerosol. Isoprene thus has important impacts for climate, air quality and human health and is the dominant chemical-biosphere-atmosphere interaction. Quantifying the magnitude and impact of these emissions is important for understanding atmospheric chemistry.

Given the importance of isoprene, long-term measurements of its concentration in tropical forests are surprisingly sparse. Here we present the first results from a long-term project to measure isoprene mixing ratios at the Bukit Atur Malaysian Global Atmosphere Watch site on the island of Borneo using both a conventional in-situ GC-FID instrument and a new smaller portable unit. Concentrations from both systems range from 0 to ~3 ppb, consistent with previous observations at the site made as part of the OP3 project. However, the shape of the diurnal variation in concentration differs from other (OP3) observations made at the same site.
We investigate the magnitude and variability of our data and the OP3 measurements using the GEOS-Chem model, which uses MEGAN version 2.1 for biogenic emissions. We find that the model significantly overestimates both isoprene concentrations and fluxes. We attribute much of this overestimation to the model emissions algorithm, which assumes tree characteristics are the same in all tropical broadleaf forests around the globe, despite the Borneo forest being dominated by dipterocarps unlike the Amazon. We update the emissions algorithm for this change and investigate the impact.

Overall we conclude that there are significant continental scale differences in the emissions of isoprene, which need to be considered when evaluating the impact of this important species.
2.011 Impacts of ozone-vegetation coupling and feedbacks on surface ozone air quality.

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Abstract:
Surface ozone is one of the most significant air pollutants due to its damaging effects not only on human health, but also on vegetation and crop productivity. Chronic ozone exposure has been shown to reduce photosynthesis and interfere with gas exchange in plants, which in turn affect the land-atmosphere exchange of energy and water, as well as surface processes that may ultimately influence the concentrations of ozone and other atmospheric constituents. Ozone damage on vegetation can thus have major ramifications on climate and atmospheric composition, including possible feedbacks onto ozone concentration itself, but the importance of such two-way ozone-vegetation coupling is not well understood. Here we examine the impacts of ozone-vegetation coupling on surface ozone through various biogeochemical and meteorological feedback mechanisms. Using the Community Earth System Model, we find that inclusion of online ozone-vegetation coupling modifies simulated ozone concentration up to +6 ppbv in China, North America and Europe, suggesting that ozone damage on vegetation constitutes a positive feedback that further enhances ozone. We find that the majority of the feedback is caused by the ozone-induced decrease in leaf stomatal conductance, which directly increases ozone by suppressing dry deposition. Reduced conductance also lowers transpiration, which increases surface temperature and thus biogenic isoprene emission, further enhancing ozone indirectly. The dry deposition-driven biogeochemical pathway and the transpiration-driven meteorological pathway contribute roughly equally to the overall positive ozone-vegetation feedback. Our work demonstrates the importance of including ozone-vegetation coupling and feedbacks in understanding and projecting future ozone air quality under climate and land cover changes over the next few decades.
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Abstract:

Currently available global and regional inventories of biomass burning emissions of gases and particles were compared over the 20th century until the year 2014. Considered datasets were created based on different approaches to emission estimation, such as historical reconstruction of burnt area, use of satellite products for burnt area, active fire data, fire radiative power, fixed or dynamical land cover and associated parameters as well as fire emission models in combination with land surface model. We compare annual totals and seasonal variation of emissions of total carbon in 14 geographical regions from the following data sets: ACCMIP, AMMABB, FINN, GFAS, GFED, GICC, MACCity, QFED and RETRO. This comparison study informs about differences among the datasets and serves as a basis for the community effort in harmonizing and creating a consistent inventory spanning through the studied period.

For most of the 20th century the inventory will need to rely on the fire emission models since the satellite observations were not available yet. We compare results of two fire emission models - mechanistic model SPITFIRE implemented in the MPI Earth system model and SiMFIRE model which is a combination of satellite-based burnt area with terrestrial ecosystem model LPJ-GUESS. We try to identify the common features and differences between the models as well as in their main driving variables.
**Abstract:**

Assessments of ozone deposition impacts on vegetation functioning is generally limited to small-scale laboratory and field experiments and some large-scale modelling studies. In this explorative study we demonstrate the potential to apply MODIS Gross Primary Production (GPP) data to assess short-term (~weeks) ecosystem scale ozone deposition impacts on vegetation functioning. Seven sites in France, Belgium, Spain and Italy were selected near measurement stations that monitor ambient ozone concentration. Multiple linear regression models were fitted to MODIS 8-day GPP data using temperature, soil moisture, evapotranspiration, land cover classes and the ozone exposure index, AOT40. The inputs were retrieved from various sources, mostly raster data with continental or global coverage of varying spatial and temporal resolution. Three land cover classes were distinguished: needle-leaved evergreen forest, broad-leaved deciduous forest and rain-fed agriculture. Thresholds beyond which short-term ozone impacts were found were set empirically for land cover classes at an 8-day AOT40 of ~250 ppb x h. A significantly negative impact of short-term ozone exposure was found beyond these thresholds resulting in an ~5% decrease in GPP decrease for an 8-day AOT40 increase of 0.1 ppm x h. Differences in sensitivity between land cover classes could not be shown due to limited amount of - as well of uncertainty in - input data, the simplicity of the used regression models and the large differences in response to ozone exposure between individual species and plants. However, despite these limitations, this study demonstrates the potential of integrating remote sensing and air quality and micrometeorological observations and model data to quantify ecosystem-scale ozone impacts on vegetation functioning.
2.016 The role of dew as a nighttime reservoir and morning source for atmospheric ammonia.

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Abstract:

Several field studies have proposed that the volatilization of NH$_3$ from evaporating dew is responsible for an early morning pulse of ammonia frequently observed in the atmospheric boundary layer. Laboratory studies conducted on synthetic dew showed that the fraction of ammonium (NH$_4^+$) released as gas-phase ammonia (NH$_3$) during evaporation is dependent on the relative abundances of anions and cations in the dew. Hence, the fraction of NH$_3$ released during dew evaporation (Frac(NH$_3$)) can be predicted given dew composition and pH. Twelve separate ambient dew samples were collected at a remote high elevation grassland site in Colorado from 28 May to 11 August, 2015. Average [NH$_4^+$] and pH were 26 μM and 5.2, respectively, and were on the lower end of dew [NH$_4^+$] and pH observations reported in the literature. Ambient dew mass (in g m$^{-2}$) was monitored with a dewmeter, which continuously measured the mass of a tray containing artificial turf to track the accumulation and evaporation of dew. Simultaneous measurements of ambient NH$_3$ indicated that a morning increase in NH$_3$ was coincident in time with dew evaporation, and that either a plateau or decrease in NH$_3$ occurred once the dew had completely evaporated. Dew composition was used to determine an average Frac(NH$_3$) of 0.94, suggesting that nearly all NH$_4^+$ is released back to the boundary layer as NH$_3$ during evaporation at this site. An average NH$_3$ emission of 6.2 ng m$^{-2}$ s$^{-1}$ during dew evaporation was calculated using total dew volume and evaporation time, and represents a significant morning flux in a non-fertilized grassland. The observed loss of NH$_3$ during nights with dew is approximately equal to the observed amount of NH$_4^+$ sequestered in dew at the onset of evaporation. Hence, there is strong evidence that dew is both a significant night-time reservoir and strong morning source of NH$_3$. 
Abstract:

Emission of biogenic hydrocarbons and the deposition of organic molecules and particles over surfaces impact the atmospheric organic carbon budget. The relative importance of upward versus downward organic carbon fluxes over forests remains observationally unconstrained. The lack of flux measurements of organic carbon is primarily due to the limited number of instruments capable of fast, sensitive and selective measurements of these compounds. During a series of field measurements at Manitou Experimental Forest in Colorado in 2015 and 2016, we used acetate chemical ionization time-of-flight mass spectrometry to make eddy covariance flux measurements of oxidized organic compounds, and an ultra high sensitivity aerosol spectrometer to make eddy covariance flux measurements of size-resolved particles. Both the potential and limitations of these techniques will be discussed. Both upward and downward fluxes of gas-phase organic compounds were observed. In addition, measurements of more reduced organic compounds through proton transfer reaction mass spectrometry provide context for the measurements. This presentation will present the flux measurements and discuss their potential impact on the atmospheric organic carbon budget.
Development, evaluation and application of a modified micrometeorological gradient method for estimating gaseous dry deposition over forest canopies.

Early Career Scientist

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Abstract:

A modified micrometeorological gradient method (MGM) was developed for estimating gaseous dry deposition velocity \( V_d \) over forest canopies. Differed from existing micrometeorological gradient methods, such as the aerodynamic gradient method (AGM) and the modified Bowen-Ratio method (MBR) which make use of concentration gradients above the canopy top, the new method uses concentration gradients between a level above and a level below the canopy top, taking advantage of relatively large gradients between these levels due to significant pollutant uptake at top layers of the canopy. The new method was validated using 10-year flux data collected at the Harvard Forest site and produced \( V_d(\text{O}_3) \) values close to the eddy-covariance measurements during daytime, although slightly overestimated the measurements at night.

The new method was further applied to a five-year gradient data collected at a forest in southern Ontario to estimate \( V_d(\text{O}_3) \) and \( V_d(\text{SO}_2) \) and produced very reasonable diurnal and seasonal patterns compared to historical and literature flux data. The mean (median) \( V_d(\text{O}_3) \) and \( V_d(\text{SO}_2) \) was 0.35 (0.27) and 0.59 (0.54) \( \text{cm s}^{-1} \), respectively at this forest. Detailed analysis of the flux data produced by this method suggests that snow surface became an effective sink for \( \text{SO}_2 \) but inhibited the \( \text{O}_3 \) deposition in winter. Canopy wetness increased the non-stomatal uptake of \( \text{O}_3 \) while decreasing the stomatal uptake, which also applied to \( \text{SO}_2 \), but additional factors such as surface acidity also played an important role on the overall uptake. Flux data at this forest produced by this new method were then used to evaluate and improve the dry deposition algorithms currently used in North America.
Impact of atmospheric light absorbing aerosol deposition on the seasonal water balance in snow dominated catchments.

Early Career Scientist

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Abstract:

The deposition of atmospheric aerosol is poorly constrained in atmospheric transport models, yet this variable has the potential to exert significant variability on the seasonal water balance in a snow dominated catchment. Light absorbing aerosols deposited on snow impact the snow melt by increasing the snow's ability to absorb short wave radiation. The consequences are a shortening of the snow duration and, on a catchment scale, a temporal shift in the discharge generation during the spring melt season. This has potential to alter ecosystem processes, especially in cold climate regions and high mountain areas, where the growing season is strongly dependent on the snow duration. To investigate the impact of Light Absorbing Impurities (LAI) on the snow pack evolution and discharge generation on a catchment scale, we use a hydrologic catchment model and have implemented a dynamic surface energy balance scheme for the snow that is forced directly from deposition rates of LAI from an atmospheric transport model.
2.021 Atmospheric Reactive Nitrogen in Rocky Mountain National Park.

Early Career Scientist

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Abstract:

Deposition of excess atmospheric reactive nitrogen in Rocky Mountain National Park (RMNP) poses threats to sensitive ecosystems. The Front Range urban corridor in Colorado, located east of RMNP, includes a variety of urban sources of nitrogen oxides, while high emissions of ammonia are found in agricultural sources on the eastern plains of Colorado. Periods of upslope (easterly) flow can transport atmospheric reactive nitrogen from the Front Range and eastern Colorado into the park. It is important to understand the degree to which these different urban and agricultural sources impact RMNP, and how meteorological conditions are associated with high-deposition periods in the park. Research associated with reactive nitrogen in the RMNP started in the mid-2000s. Ongoing measurements at RMNP include 24hr-integrated gaseous and particulate nitrogen species measured by denuder/filter pack methods, gaseous ammonia measured continuously by cavity ring-down spectroscopy (CRDS), and wet deposition of nitrogen compounds. Measurements of gaseous ammonia at high-time resolution are also available at sites to the east in Greeley and Loveland, CO. The combination of these three sites with high-time resolution ammonia concentrations helps document westward transport of ammonia from NE Colorado into RMNP. A small ensemble of forecasts using the Advanced Research version of the Weather Research and Forecasting (WRF-ARW) model has been run routinely to calculate air parcel trajectories released from the eastern plains of Colorado to RMNP. An Early Warning System allows agricultural producers to implement management practices that reduce nitrogen emissions when an upslope event is forecast. Early Warning System predictions will be evaluated in this presentation to examine how well the model predicts high concentrations of ammonia and high nitrogen deposition events in RMNP.
Investigating Ammonia Sources with the Cross-Track Infrared Spectrometer (CrIS).

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Abstract:
Ammonia (NH$_3$) can react with sulfuric and nitric acid in the atmosphere to produce ammonium nitrate and ammonium sulfate aerosols. Uncertainty in NH$_3$ emissions, primarily from agricultural sources such as animal feed lots and fertilizer use, can thus lead to uncertainty in the formation, vertical distribution, and radiative impacts of ammonium nitrate and ammonium sulfate aerosol, which in turn can lead to significant uncertainties in air quality and climate models. Here we present the first use of the Cross-Track Infrared Sounder (CrIS) aboard the Suomi NPP satellite to investigate and improve modeled concentrations of NH$_3$ in California from the Community Multiscale Air Quality (CMAQ) model. Our baseline model simulation uses the bi-directional NH$_3$ flux capability of CMAQ along with a version of the California Air Resources Board (CARB) emission inventory that includes an empirically derived diurnal cycle for NH$_3$ emissions. The extensive spatial and temporal coverage of the CrIS satellite provides a wealth of data on NH$_3$ concentrations over California. We will discuss the errors identified in our emission inventory as well as the meteorological and other conditions leading to remaining discrepancies between the model and observations. We will also present recommendations for the further use of CrIS data in atmospheric chemistry studies.
Biomass burning smoke predictions across scales: from regional forecasts using near-real-time emission constraints to city scale simulation and inversion of a fire plume.

Early Career Scientist

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Abstract:

Smoke from biomass burning is a global scale pollutant generating deep impacts to our society and planet. Thus, advancements in our ability to model and forecast this phenomena is crucial to further our understanding of the impacts. In this presentation we will show new developments and findings on multi-scale smoke simulations. First, we will present a novel system to operationally forecast smoke that includes near-real-time emission constraints based on aerosol optical depth (AOD) observations from ground-based and satellite data. We will show the performance of such system on the smoke forecasts from Central Africa, smoke transport over the Southeast Atlantic and smoke impact on the persistent stratocumulus deck in preparation for and during the NASA ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) field experiment. Second, we will present results of full-chemistry simulations at 2km resolution for a case study over Chile, where smoke from a wildfire heavily affected the air quality in the city of Santiago for about a week. The impacts from the fire were observed with multiple measurements, including an air quality network, ground-based Lidar, an AERONET site and 1km resolution satellite-based AOD retrievals (MAIAC algorithm), which can enable the evaluation of the model’s ability to represent smoke loads and spatial (vertical and horizontal) distributions. We further apply a variational
inversion technique to better constrain the fire emissions with these observations, never done before at this scale. The results provide key information to understand the underlying biases and how to improve them. Finally, preliminary findings from the KORUS-AQ (Korea-US Air Quality study) field experiment with respect to fires from agricultural burning in China and wildfires in southern Siberia will be briefly presented.
Abstract:

Gases from both anthropogenic and biogenic sources have the potential to have a profound impact human health and climate. Emissions from biomass burning include HCN, HCHO, NH₃, and C₂H₆. We describe the University of Colorado mobile Solar Occultation Flux (CU mobile SOF) instrument to quantify sources of emissions in the UV-Visible and Infrared wavelengths. The SOF method is complementary to in-situ observations in being able to obtain trace gas fluxes and trace gas production rates from a source when driving around or upwind and downwind of that target source. The Volkamer group has developed a digital mobile solar tracker (Baidar et al., 2016, AMT) that can be simultaneously coupled to a Fourier Transform Infrared Spectrometer (FTIR) and a UV-Visible Spectrometer (UV-Vis) to measure trace gases along the direct solar beam.

The mobile lab was successfully deployed during FRAPPE (Front Range Air Pollution and Photochemistry Experiment) in Colorado in 2014, and the CU mobile SOF instrument was successfully used to map and quantify emission fluxes from urban areas (e.g. NO₂), oil and natural gas operations (C₂H₆) and feedlots (NH₃). The total column measurements were conducted at high spatial (5-19 m) and temporal (2 sec) resolution. We measured significant variability in the gas columns.
During the summer of 2016 the CU mobile SOF will be deployed on an aircraft. Due to the often elevated fire plumes in biomass burning events, and the size and long-range distances that these plumes travel in the atmosphere, instrumentation that are capable of operating on aircraft are a necessary component to field experiments that study fires. We present results from select research drives from FRAPPE and pending progress will report first results from the airborne deployment of the mobile SOF.
2.026 Emissions of CO2, CO, and CH4 from peat forest fires on Sumatra Island in non El-Niño year 2013.

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Abstract:

We observed substantial enhancements of atmospheric trace gases and aerosols (CO2, CH4, CO, PM2.5, PM10, and black carbon) in summer of 2013, with continuous instruments onboard the NIES voluntary observing ships sailing in the Southeast Asia region. The enhancements were observed off the east coast of the Malay Peninsula and in the straits of Malacca along the shipping route, associated with prominent enhancements in CO. The 6-year climatology (2008 – 2013) of the monthly mean CO mixing ratios in these areas shows the maximum in June, followed by moderate but relatively high values in following months of July and August. Our analysis combining in-situ measurements, satellite observations, and an air trajectory analysis showed that the observed enhancements were due mainly to the intensive biomass burnings that occurred in the central Sumatra. We examined the quantitative relationships between the observed CO2, CO, and CH4 during the CO enhancement events. Strong correlation was found between these gases, and the calculated emission ratios of CO/CO2 suggested large contribution of peat forest fires to the observed enhancements (CO/CO2 = 135 ppb/ppm). We determined the emission factors (EF) of CO2, CO, and CH4 from the peat forest fires using the emission ratios of CO/CO2 and CH4/CO2, showing the EFs of 1722, 132.0, and 6.27 (g/kg) for CO2, CO, and CH4, respectively. These EFs were compared with those calculated from the current emission inventory (GFED version 4S), which are 1671, 144.5, and 11.95 (g/kg) for CO2, CO, and CH4 respectively. These EFs were consistent with each other except CH4, suggesting that the EF used in GFED4S is overestimated for CH4 from peat fires.
**2.027 Temperature and burning history affect emissions of greenhouse gasses and aerosol particles from tropical peatland fire.**

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Abstract:

Peatland burning is a major source of greenhouse gasses as well as atmospheric trace species such as aerosol particles. Burning of tropical peatland in equatorial Asia has intensified over the last decades, emitting huge plumes of haze that can blanket large parts of Southeast Asia. Both laboratory and field studies have previously been conducted to investigate emission from peat burning, yet a significant variability in data still exists. We conducted a series of experiments to characterize the gas and particulate matter emitted during burning of a peat samples from Sumatra in Indonesia. Heating temperature of peat was found to regulate the ratio of $\text{CH}_4$ to $\text{CO}_2$ in emissions ($\text{CH}_4/\text{CO}_2$) as well as the chemical composition of particulate matter. The $\text{CH}_4/\text{CO}_2$ ratio was larger for higher temperatures, meaning that $\text{CH}_4$ emission is more pronounced at these conditions. Mass spectrometric analysis of organic components indicated that aerosol particles emitted at higher temperatures had more unsaturated bonds and ring structures than that emitted from cooler fires. In addition, $\text{CH}_4$ emitted by burning charcoal derived from previously burned peat was lower by at least an order of magnitude than that from fresh peat. These results highlight the importance of both fire history and combustion temperature for the composition of tropical peat-fire emissions. They suggest that remote sensing technologies that map fire histories and temperatures could provide improved estimates of emissions.
2.029 CANEXMIP: Intercomparison of models for simulating canopy-atmosphere exchange and chemistry of reactive compounds and aerosols.

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Abstract:

The so-called “big-leaf” approach has typically been used to represent surface emissions and deposition of reactive compounds and aerosols in large-scale atmospheric chemistry models. This has been a reasonable approach for studies focused on acidification and eutrophication research but consideration of the role of more reactive compounds, that undergo fast chemical transformation at timescales comparable to turbulent transport and deposition, requires the explicit consideration of interactions between emitted and depositing compounds within the canopy airspace. Consideration of canopy interactions is also relevant for assessing the efficiency of the release of emitted reactive compounds (and products) as well as for comparison of model simulated surface layer concentrations with observations. Temporal variability in these surface layer concentrations is largely determined by the canopy interactions that are not explicitly captured by the “big-leaf” approach. In addition, to assess the potential impact of pollutant deposition on ecosystem functioning, the role of canopy interactions in determining stomatal versus non-stomatal deposition must be considered. There is a suite of models that explicitly consider these canopy interactions and are applied mostly for field-scale analysis of concentration and flux measurements. In addition, there are also ongoing efforts to improve multi-layer canopy exchange models for incorporation in large-scale models. These models are now being compared and assessed by the joint iLEAPS (integrated Land Ecosystem-Atmosphere Processes Study) -GEIA (Global Emission Inventory Activity) Canopy EXchange Model Intercomparison Project (CANEXMIP). We will present the rationale for the CANEXMIP activity and provide an overview of studies that have focused on these canopy interactions. In addition, we will present more details on the activities and initial intercomparison of canopy exchange models with tropical forest observations. Plans for assessing the model skills for simulating reactive trace gas and aerosol exchange will be discussed and needs for field observations will be described.
2.030 Drought impacts on high ozone in California.

Early Career Scientist

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Abstract:

California is currently experiencing the most severe drought both since the start of the 120-year observational record and over the last millennium. California is also home to the worst ozone (O₃) air quality in the U.S., with the top 5 most O₃-polluted cities in the U.S. located in the state. Here, we explore the drought’s influence on high O₃ in the polluted agricultural region of California’s San Joaquin Valley. We describe drought effects on O₃ production chemistry, on O₃ stagnation over multiple days, and on the O₃ lifetime. We also consider the atmospheric implications of the adaptive agricultural practices taking in place in the region. Climate change has been implicated as a driver of the severity of the current drought in California, suggesting that the response of O₃ to the 2012-2015 drought may offer insight into air quality in the future.
Non-stomatal uptake controls inter-annual variability in ozone dry deposition velocity over a northern mid-latitude deciduous forest.

Early Career Scientist

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Abstract:

Our understanding of ozone removal by northern mid-latitude broadleaf deciduous forests is largely based on short-term observational studies. Year-to-year variations of this sink have received little attention. Given the importance of ozone dry deposition to the tropospheric ozone budget and regional air quality, an improved mechanistic understanding of this sink is needed to model ozone accurately. We investigate inter-annual variability (IAV) in seasonal and diel cycles of ozone dry deposition velocity (vd,O3) using nine years of hourly ozone eddy covariance measurements at Harvard Forest, a deciduous forest in central Massachusetts, USA. Coincident water vapor and carbon dioxide eddy covariance and micrometeorological measurements enable us to examine ozone dry deposition in the resistance-in-series framework and to estimate stomatal conductance (g_s) via two independent methods. For all months of the year, monthly daytime mean vD,O3 at Harvard Forest during the lowest vs. highest year differs by approximately a factor of two. These year-to-year differences are not apparent in nine years of simulated vD,O3 by a state-of-the-art chemistry-transport model with a modified Wesely scheme and driven by observed meteorology. We find that canopy resistance determines IAV in vD,O3 during the growing season at Harvard Forest. While the two g_s estimates disagree in magnitude and in the shape of the diurnal cycle, both estimates indicate little IAV compared to total canopy uptake, and a similar ranking of years with low vs. high g_s. We conclude that non-stomatal uptake is the dominant driver of the observed IAV. Some years have consecutive months with high (or low) vD,O3 (and thus non-stomatal uptake), implying that seasonal environmental conditions influence the non-stomatal uptake of ozone. Once identified, these environmental controls could be incorporated into Wesely-based schemes in global models, improving the representation of IAV in this key ozone sink.
**2.032 Development of top-down emission inventories and models for non-anthropogenic sources; examples of vegetation fires, biogenic aerosols, sea salt, and wind-blown dust.**

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Abstract:  
Top-down emission estimation via inverse dispersion modelling can be used for problems, where bottom-up approaches are difficult or uncertain. Emission from wild-land fires, biogenic aerosols from vegetation, wind-blown dust and sea salt, all strongly depend on environmental forcing and ecosystem state, which limits the value of static emission inventories. In combination with dispersion modelling, the satellite and/or in-situ observations can be used to constrain the time- and space-resolving emission fields. The approach then refines the a-priori emission (inventories, lab studies, etc) for real-life situations using the inverse-modelling technique.  
The emission calculation includes two steps:  

- top-down calibration of emission factors and model parameters via inverse problem solution that is made once using training dataset from the past,  
- bottom-up application of the obtained emission model in dispersion computations using the appropriate input data, such as individual-fire radiative energy observations.

This procedure can be extended with a dynamic adjustment of the emission model via assimilation of available observations.  
However, the approach also has significant uncertainties. One of them refers to inaccuracies of the inverse problem solution that originate from imperfect observations, shortcuts in the model formulations and assimilation algorithm, etc. Using examples of the SILAM CTM applications to several fire episodes, and vegetation blossoming and dust outbreaks, it is pointed out that the top-down system calibration performed for a limited number of comparatively moderate cases (often the best-observed ones) may lead to errors in application to extreme events.  
Our experience also showed that “second-class” effects, which are usually ignored or simplified, can easily bring a few times difference in the model-measurement comparison: sea surface temperature and salinity for sea-salt emission, fire aerosol size distribution, diurnal variation and injection height, sand size and composition in deserts, vegetation treatment and anthropogenic stress on plants, are among the most-significant factors.
2.033 African Dust as a source of nutrients to a Tropical Montane Cloud Forest in the Caribbean.

Early Career Scientist

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Abstract:

Huge amounts of African dust travels thousands of kilometers from the Sahara and Sahel regions to the Caribbean, northern South America and southern North America. These dust particles can play an important role in cloud formation and be deposited to the ecosystems as wet deposition in cloud and rainwater. In order to improve our understanding of the role of long-range transported African dust (LRTAD) as a source of nutrients in a tropical montane cloud forest (TMCF) in Puerto Rico, we had field campaigns measuring dust physical and chemical properties in summers of 2013, 2014 and 2015, as part of the Luquillo Critical Zone Observatory (LCZO). Measurements were performed at the TMCF of Pico del Este (PE, 1051 masl) and at the nature reserve of Cabezas de San Juan (CSJ, 60 masl), that serves as a control station. In both stations we monitored meteorological parameters (e.g., temperature, wind speed, wind direction). At CSJ, we measured light absorption and scattering at three wavelengths (467, 528 and 652 nm). At PE we collected cloud and rainwater for chemical analyses. Samples were classified as low or high dust influenced using data from models, satellites and CSJ measurements. Soluble ions, insoluble trace metals, pH, conductivity, total and dissolved organic carbon and nitrogen were measured for cloud and rainwater. Enrichment factor analysis was used to determine sea and crustal contribution of species by sample, as well as the neutralization factor and fractional acidity. Some preliminary results show cloud water conductivity for low and high dust periods was 47.7 vs 81.1 uS/cm, respectively, and for rainwater was 12.8 vs 15.0 uS/cm. pH showed differences no larger than 11% for both 2013 and 2014 periods. Also, increases in the overall ion concentration were seen in high dust samples. Detailed results will be presented at the meeting.
**Abstract:**

Surface ozone (O$_3$) is an air pollutant and greenhouse gas that is toxic to plants, reducing their growth and ability to regulate water loss. Past controlled, single-species experiments have shown that current atmospheric O$_3$ levels degrade water-use efficiency (WUE), which is the ratio of carbon uptake in photosynthesis to water loss in transpiration, in several crop and tree species. This implies that O$_3$ air pollution modifies the terrestrial water cycle and precipitation, but no prior studies have documented the O$_3$ impact on WUE at the ecosystem level.

We correlate WUE with stomatal O$_3$ uptake and meteorological factors at 23 eddy-covariance flux towers in the US and Europe that span a wide range of temperate forest and crop ecosystems. After removing the mean seasonal cycle of all variables, we find that daily anomalies in stomatal O$_3$ uptake degrade WUE by 1-3% at most sites ($p < 0.05$), in addition to the expected dependencies on temperature, humidity, and photosynthetically active radiation (PAR). This O$_3$ impact on WUE is similar to that found in controlled, single-species experiments and of the same magnitude as the response to daily variations in temperature, humidity and PAR. Affected ecosystems include broadleaf crops and all forest types, indicating that O$_3$ impacts are widespread among many species that have not been studied individually and that the aggregated effects across an ecosystem are similar to those of individual species. The largest impacts on WUE occur in ecosystems with high stomatal conductance, such as broadleaf forests, humid climates, or irrigated crops, rather than where surface O$_3$ concentrations are highest. The weakest O$_3$ impacts occur in ecosystems with large populations of C4 plants, but longer data records are needed to confirm this at the ecosystem level. Ongoing work is exploring how well land biosphere models can simulate these effects of O$_3$. 

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**2.034 Ozone deposition degrades water-use efficiency across multiple ecosystems.**

Early Career Scientist

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2.038 The effects of canopy mixing on fluxes and vertical concentration gradient of VOCs above a forest canopy.

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Abstract:

Fluxes of biogenic Volatile Organic Compounds from forest ecosystems account for over three-quarters of the hydrocarbons annually released to the atmosphere. The atmospheric reactions of these compounds affect the composition, chemistry and oxidative capacity of the troposphere on all time and spatial scales. They are well-documented precursors of ozone and aerosol as well as a source of reactive nitrogen to remote regions. In spite of recent advances in our understanding of their reactions across a spectrum of NOx regimes, models are often still unable to reconcile simulated concentrations and fluxes of bVOCs with those measured in and above forest canopies, particularly in regions where NOx concentrations are moderate (~1-2 ppbv). We apply the FORCaST (FOREst Canopy Atmosphere Transfer) canopy exchange model to a rural mixed deciduous forest site in the northern mid-latitudes where NOx mixing ratios are typically around this level. We explore the canopy processes controlling exchanges of isoprene and its oxidation products methyl vinyl ketone and methacrolein between the forest canopy and the atmosphere. While isoprene fluxes are always positive, bi-directional exchange of methyl vinyl ketone and methacrolein has been observed at many sites. Furthermore, the chemical lifetime of isoprene is of a similar order to the canopy retention time making vertical exchange processes important. We conclude that for such species understanding and better accounting for turbulent mixing is as important as chemistry in determining canopy-top fluxes.
2.039 Assessing the role of dry deposition in observed ozone-meteorology correlations.

Early Career Scientist

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Abstract:

Strong seasonally and spatially dependent correlations between surface ozone (O3) and meteorological variables have been reported by many authors and implemented in phenomenological air quality forecast models. While the meteorological dependence on O3 production (PO3) is well known, the strength of site specific O3-meteorology correlations is not always well captured by mechanistic models. Chemical transport models (CTMs) are unable to accurately predict the strength and location of the ozone-relative humidity negative correlation seen widely in summer time observations with differing explanations given for this failing.

Using 20 years of hourly O3 and meteorological data from the Environmental Protection Agency’s Clean Air Status and Trends Network (CASTNET) and a coupled atmosphere-biosphere box model, we look at the role of individual ozone production and loss processes in regulating O3 concentrations as a function of
meteorology. We find that it is only with the inclusion of an ozone deposition scheme with a fully meteorologically dependent stomatal uptake parametrization that we can explain the strength, seasonality, and spatial dependence of the ozone-relative humidity correlation seen in CASTNET observations. Deposition to vegetation is responsible for a significant portion of ozone loss in the continental surface layer during the growing season and as such needs to be treated accurately. This poses a challenge to CTMs run at coarse resolutions with limited land-use data and simplified treatment of stomatal uptake, and as such we should not expect these models to predict the ozone-relative humidity correlation seen in real data.
2.040 Regional air quality degradation in Northern South America due to biomass burning transboundary pollution.

Early Career Scientist

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Abstract:

The Orinoco river basin is a 98-million hectares (Mha) savanna ecosystem shared by Colombia and Venezuela, of which 35 Mha are located in Colombia. The savanna ecosystem undergoes periodic biomass burning, particularly during the dry season. It is estimated that on average ~3 Mha are burned per year only in the Colombian side. Fire activity in the Orinoco region can be natural or human induced. The latter includes among other causes the pest control and the use of fire for pasture management due to the low palatability of native pastures, which is improved in the fresh regrowth. By April, the raining season starts in Colombian llanos and fire activity is reduced. However, in Venezuelan llanos there is still high fire activity during April and May. These differences on period and duration of dry seasons imply that biomass burning plumes from one country can be transported to the other. The environmental authority of Arauca and Yopal (two cities located at the Colombian Orinoco Foothills) made particulate matter measurements during April and May 2015. We will present the analysis of particulate matter levels in those cities and their relation with biomass burning in Venezuelan Orinoco savannas. Concentrations of PM10 were considered high taking account that are small cities with little industrial activity and little mobile sources. There were some concentrations over the daily air quality standards of Colombia (100μg/m³). Both the levels and tendencies of PM10 were similar for both cities, which suggest a regional effect of pollution sources. Back trajectories simulations using STILT model showed that for most of the sampling days, air masses arriving to Arauca and Yopal came from Venezuela. High correlation was found between the PM10 average concentration and fire activity in Venezuela, which evidenced the existence of a transboundary pollution problem.
2.042 Using mobile laboratory and aircraft measurements to characterize feedlot emissions and their contribution to atmospheric methane over the Denver-Julesburg Basin.

Early Career Scientist

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Abstract:

Atmospheric emissions from animal husbandry are important to air quality and climate, but are hard to characterize and quantify as they vary substantially based on management practices, livestock type, and diurnal and seasonal cycles. Using a new mobile laboratory, ammonia, methane, nitrous oxide, and carbon dioxide emissions were measured from several concentrated animal feeding operations (CAFOs) in northeastern Colorado. Four CAFOs were chosen for repeated diurnal and seasonal measurements. A diurnal trend in the enhancement ratio of ammonia to the other compounds is clearly observed and is consistent across seasons and CAFOs. These findings are used to develop a source signature for feeding operations in the area.

In addition to 250+ CAFOs, the Denver-Julesburg basin (DJB) is a heavily developed oil and natural gas region with over 25,000 wells and numerous compressors and processing plants. Due to the co-location of these varied methane point sources, top-down measurements are often unable to attribute emissions to a specific source or sector. In this work, the CAFO emission signature determined from targeted mobile laboratory measurements is combined with aircraft measurements of ammonia, methane, and ethane during the spring 2015 Shale Oil and Natural Gas Nexus (SONGNEX) field campaign to attribute atmospheric methane over the DJB to either agriculture or fossil fuel sectors.
Properties and impacts of biomass burning aerosol over the Amazon region - a summary of the South American Biomass Burning Analyses (SAMBBA) project.

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Biomass burning from wildfires is one of the largest sources of absorbing and organic aerosol on the planet. It has a significant influence on cloud properties and makes a substantial contribution to the radiation balance of the atmosphere on continental scales. Such perturbations have an effect on regional weather and climate and can influence net growth of the biosphere. Despite the importance of these effects across continental regions such as Amazonia, there are considerable gaps in our knowledge that limit accurate prediction. These result from: inaccurate prediction of optical properties from the physical and chemical properties of biomass burning aerosol; uncertainty over the controls on the aerosol distribution throughout the atmosphere resulting from emissions and dispersion of large plumes; the inability of regional and global models to represent the measured atmospheric burden across major regions of burning without significant moderation of emissions; recent changes in burning practices that significantly alter the optical properties and spatial extent of biomass burning aerosol; shortcomings in model representations of absorbing aerosol that limit our understanding of aerosol-cloud-precipitation feedbacks; and the complexity of capturing the net response of the biosphere to the competing stimuli of reduced radiation, increased fraction of scattered light, and increased ozone resulting from biomass burning.

During 2012 a major experimental study to investigate biomass burning across the southern margins of Amazonia took place. We present the major results from this joint Brazil-UK programme, which involved a large research aircraft – the UK Atmospheric Research Aircraft (FAAM) and a number of ground-based measurement sites, and utilised a range of regional and climate models. We demonstrate how this study has been able to address or constrain the process uncertainties discussed above, advance methods to capture feedbacks and quantify their magnitude, and summarise the remaining outstanding research questions.
Exploring the direct impacts of particulate matter and surface ozone on global crop production.

Early Career Scientist

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Abstract:

The current era of rising food demand to feed an increasing population along with expansion of industrialization throughout the globe has been accompanied by deteriorating air quality and an enhancement in agricultural activity. Both air quality and the food supply are vitally important to sustaining human enterprise, and understanding the effects air quality may have on agricultural production is critical. Particulate matter (PM) in the atmosphere decreases the total photosynthetically available radiation (PAR) available to crops through the scattering and absorption of radiation while also increasing the diffuse fraction (DF) of this PAR. Since plants respond positively to a higher DF through the more even distribution of photons to all leaves, the net effect of PM on crop production depends on the magnitudes of these values and the response mechanisms of a specific crop. In contrast, atmospheric ozone always acts to decrease crop production through its phytotoxic properties. While the relationships between ozone and crop production have been readily studied, the effects of PM on crop production and their relative importance compared to ozone is much more uncertain. This study uses the GEOS-Chem chemical transport model linked to the RRTMG radiative transfer model and the DSSAT crop model to explore the impacts of PM and ozone on the globally distributed production of maize, rice, wheat and soybeans. First, we examine how air quality differentially affects total seasonal production by crop and region. Second, we investigate the dependence of simulated production on air quality over different timescales and under varying cloud conditions.
2.045 Contribution of post-harvest agricultural paddy residue fires in the N.W. Indo-Gangetic Plain to ambient carcinogenic benzenoids, toxic isocyanic acid and carbon monoxide.

Early Career Scientist

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Abstract:

In the North West Indo-Gangetic Plain (N.W.IGP), large scale post-harvest paddy residue fires occur every year during the months of October–November perturbing the regional air quality posing health risks for the population exposed to high concentrations of carcinogens such as benzene and toxic VOCs such as isocyanic acid. These gases and carbon monoxide are known to be emitted from biomass fires along with acetonitrile. Yet no long-term in-situ measurements quantifying the impact of this activity have been carried out in the N.W. IGP. Using real time in-situ measurements of these gases at a strategic downwind site over a three year period (2012–2014), we demonstrate the strong impact of this activity on their ambient concentrations. In contrast to the pre-paddy harvest period, excellent correlation of benzenoids, isocyanic acid and CO with acetonitrile (a biomass burning tracer); \( r \geq 0.82 \), distinct VOC/acetonitrile emission ratios and high ambient concentrations of these species were observed during the post-paddy harvest period. The average concentrations of acetonitrile, benzenoids and CO in the post-paddy harvest periods were about 1.5 times higher than the annual average concentrations. Moreover, annual and post-harvest concentrations of isocyanic acid is close to 1ppb, the concentration considered to be sufficient to enhance risks for cardiovascular diseases and cataracts. The annual average concentrations of benzene (class A carcinogen), exceeded the annual exposure limit of 1.6 ppb at NTP. We show that mitigating the post-harvest paddy residue fires can lower the annual average concentration of benzene and ensure compliance with the National Air Quality Standard. Calculations of excessive lifetime cancer risk due to benzene amount to 25 and 10 per million inhabitants for children and adults, respectively, exceeding the USEPA threshold of 1 per million. Thus, there is an urgent need for enforcement/implementation of economically viable solutions to mitigate the paddy residue fires.
Tracking agricultural soil NOx and NH3 emissions variability with novel methodologies.

Early Career Scientist

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Abstract:

Agricultural production systems significantly perturb the reactive nitrogen cycle via significant atmospheric nitrogen oxides (NOX) and ammonia (NH3) emissions. NOX and NH3 serve as precursors to ozone and ammonium nitrate aerosols, linking agriculture, air quality, radiative forcing, and ecosystem health. Constraining agricultural NOX and NH3 emissions is critical for closing agro-ecosystem nitrogen budgets. However, fluxes are highly uncertain and lack widespread, high-resolution measurements for capturing spatially heterogeneous soil emission pulses, especially on diurnal timescales. We characterize NOX and NH3 fluxes and the nitrogen isotopic composition of NOX from cropland soils across a variety of representative fertilizer and water management scenarios. A field and laboratory-verified technique for actively capturing NOX in solution is optimized for hourly resolution soil NOX isotopic measurements. We evaluate the ability of nitrogen isotopic enrichment factors of NOX emissions relative to inorganic soil nitrogen substrates to distinguish emission processes. We also demonstrate a novel NH3 flux chamber measurement method, using a portable, battery-powered, open-path quantum cascade laser-based NH3 sensor, that accounts for adsorption losses to chamber surfaces. Ammonia detection from 50 ppbv to 50 ppmv is optimized for a large dynamic range and several minute resolution flux measurements. We present in-situ soil
flux studies with soils and fertilizers representative of a sustainable cropping system of no-till and manure injection in Pennsylvania and an irrigated cropping system with conventional and no-till in Colorado. Continuous diurnal flux measurements are performed to investigate correlations of soil NO$_x$ and NH$_3$ fluxes with fertilizer application, response to rainfall/irrigation, and diurnal soil temperature variations. Soil surface-applied manure resulted in order of magnitude higher NH$_3$ fluxes than below surface incorporation. These observations have implications for future agricultural management and mitigation strategies and are applicable for comparisons with field-scale eddy flux observations and validations of satellite NO$_2$ and NH$_3$ model inversions in agricultural emission regions.
2.049 The impact of extreme vegetation fires in South East Asia.

Early Career Scientist

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Abstract:

Illegal vegetation fires have been estimated to cause approximately 11000 premature deaths during intense fire years in highly densely populated regions in South-East Asia. Strong El-Niño and positive Indian Ocean Dipole conditions are associated with an increase in the frequency and intensity of vegetation fires in Indonesia and Borneo, enhancing population exposure to hazardous concentrations of smoke and air pollutants. In this work we investigate the impact on human health and climate of the vegetation fires in South East Asia during Fall 2015, which were the largest of the past two decades. We performed high resolution simulations using the Weather Research and Forecasting model with coupled Chemistry based on a new release of fire emissions from the Fire INventory from NCAR and a sectional approach for aerosol size distribution. Model skill is
evaluated against a suite of space- and ground-based observations of aerosol optical properties and concentrations of particles and trace gases. The model is able to capture both the spatial and temporal variability of extreme pollution episodes and allows for identification of the sources and transport of pollutants over South East Asia. The impact of these events on climate and human health is quantified relative to a control run without fire emissions and by integrating high resolution population density maps with concentration response functions and threshold exceedances analysis. Results from this research provide decision-relevant information to policy makers regarding the impacts of land use changes and human driven deforestation on fire frequency and population exposure to degraded air quality.
The development of modelling methods to assess the combined threat of climate extremes and ozone on ecosystems.

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Abstract:

Extreme climate and ground level ozone (O₃) air pollution stress are likely to co-occur and affect ecosystems. This is due to elevated O₃ episodes being more frequent under hot, dry sunny conditions as well as in rural agricultural regions (downwind of source O₃ precursor pollutant emissions). Most pollution risk assessment studies have used methods that relate damage to ambient ozone (O₃) concentrations rather than stomatal O₃ flux, now widely accepted as the most suitable predictor of damage. Even where stomatal O₃ flux is used, studies rely on whole season accumulations from which to determine yield losses even though O₃ will be compromising photosynthetic capacity over shorter time-periods (hours to days) as well as over whole growing seasons.
Advances in climate modelling provide opportunities to use a combination of finer spatially and temporally resolved meteorological data (for both current and future projected climates) in conjunction with a new photosynthetic based O₃ deposition and stomatal flux model (DO₃SE) to produce novel methods to assess the effects of interactions between nitrogen availability (soil fertility), heat, drought and O₃ on photosynthesis, crop growth and yield. These efforts will focus on regions in Asia, where high O₃ concentrations and climate extremes are already threatening crop productivity in a food insecure region. These new risk assessment methods will be able to inform policy through evaluating a number of emission storylines to identify those most likely to mitigate the effects of both O₃ pollution and climate change. The work will also develop new O₃ damage crop modelling methods that can be easily incorporated into existing photosynthesis-based crop modelling methods for application among the wider crop modelling community.
Forests cover over 30% of the Earth’s land surface and play a key role in the global climate. Extensive land-use change has occurred over the past few decades, converting forests to agriculture, pasture and urban areas. This land-use change impacts global climate through changes to surface albedo (biophysical impacts) and through altering emissions of carbon dioxide and other trace gases and particles (biogeochemical impacts). Both the biophysical and biogeochemical impacts of land-use change are poorly constrained. We combine data from different satellites to quantify the impacts of land-use change on surface albedo and atmospheric composition. Using data from the Landsat satellite, we identify regions of land-use change over the period 2000 to 2014. Using data collected by the Moderate-Resolution Imaging Spectroradiometer (MODIS) instrument and Ozone Monitoring Instrument (OMI) we assess the impacts of forest cover change on surface albedo, aerosol optical depth (AOD) and NO₂. The results from these findings will act as a base to evaluate the effectiveness of the coupled chemistry climate model, UKCA, to capture biogeochemical and biophysical processes associated with land use change.
Ambient concentrations of BVOCs at a South African grazed grass-savannah ecosystem.

Early Career Scientist

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Abstract:

Ecosystems of the earth produce and emit various biogenic volatile organic compounds (BVOCs), which are involved in plant growth and reproduction, as well as acting as defensive compounds for plants. In an ecosystem the BVOC production rate depends on several physical (e.g. temperature, precipitation, moisture, solar radiation and CO₂ concentration) and biological parameters (e.g. plant species, plant-specific emission capacity, phenology, biotic and abiotic stresses, attraction of pollinators); typically 0.2-10% of the carbon uptake in photosynthesis is converted to BVOCs. BVOCs are closely associated with atmospheric chemistry in the troposphere, e.g. in the formation of...
secondary pollutants such as ozone and secondary organic aerosols (SOA). This makes the atmospheric BVOC concentrations important input parameters for effective air quality management, as well as for the modelling of atmospheric chemistry at global, regional and local scales.

Despite the significance of atmospheric BVOCs, the knowledge on BVOCs in southern Africa is limited. Previous research in the region focused on short measurement campaigns on BVOC emission rates. The hypothesis in this study was that frequent, BVOC concentration measurements throughout an extended period can reveal changes in BVOC exchange from an ecosystem at seasonal and inter-annual time scales, which will be particularly useful in estimating the total regional VOC emissions. We report the ambient concentrations of BVOCs measured for more than two years at Welgegund (www.welgegund.org, 26.57°S, 26.94°E, 1480 m a.s.l.) measurement station located in a grazed grass savannah environment, ~100 km south-west of the Johannesburg-Pretoria megacity. To our knowledge, this is the first data set of ambient BVOC concentrations that covers more than two full seasonal cycles in southern Africa, and it indicates strong seasonal and inter-annual variability in the atmospheric BVOC levels.
2.066 Rapidly changing interactions between forests and atmospheric chemistry: contemporary changes in land cover and anthropogenic emissions.

Early Career Scientist

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Abstract:

Forests are well-known to have a large impact on atmospheric chemistry due to reactive biogenic emissions and removal by dry deposition. Few studies have explored decadal-scale changes in forest-atmosphere interactions, despite these timescales being highly relevant to contemporary changes in land cover and anthropogenic emissions. Here we discuss work that combines satellite observations with chemical transport modeling to investigate changes on this time scale and better predict air quality, climate, and ecosystem impacts. First, using global observations of NO$_2$ from multiple satellite instruments, we constrain estimates of oxidized nitrogen (NO$_y$) deposition to the world’s forests over the past twenty years. For example, we find dramatic decreases in NO$_y$ deposition to forests in the US, supporting other evidence that critical nitrogen loadings across the country are increasingly dominated by reduced nitrogen. Concurrent with changes in anthropogenic emissions are changes in forest cover due to natural and human-driven processes, which we investigate using a custom land cover module coupled to a chemical transport model. For example, we show how projected insect-driven tree mortality in the US over the next 15 years might not only impact the removal of O$_3$ by deposition, but also lead to shifts in chemical production of O$_3$ and organic aerosol due to decreases in biogenic emissions. We also use an up-to-date satellite-derived description of land cover in Southeast Asia to explore how conversion of forests to oil palm plantations might be impacting atmospheric chemistry in that region. We find that predictions of regional deposition and air quality can be very sensitive to all these changes in forest-atmosphere interactions. Anthropogenic emissions and land cover changes should both be considered in concert for a better representation of future atmospheric chemistry.
2.067 There’s the smoke, where’s the fire? A regional analysis of which fire-prone regions have the largest impact on U.S. air quality.

Early Career Scientist

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Abstract:

Wildfires represent a major challenge for air quality managers, as they are large sources of particulate matter (PM) and ozone precursors. Smoke can be transported thousands of kilometers to deteriorate air quality over large regions downwind. Under a warming climate, fire severity and frequency are likely to increase, exacerbating an existing problem. Using the National Environmental Satellite, Data and Information Service (NESDIS) Hazard Mapping System (HMS) smoke data, we identify fires that are confirmed to have produced smoke over the U.S. and Canada between 2005-2015. These smoke producing fires are the fires used to initiate the National Weather Service smoke forecast. We use these fires combined with Hybrid Single Particle Lagragian Integrated Trajectory Model (HYSPLIT) forward trajectories, satellite detected smoke plume data, and detailed land use data to develop a climatology of the land type, ownership (public versus private), location, and seasonality of the smoke that affects each of the 9 continental Environmental Protection Agency (EPA) regions. This work offers a present-day look at the spatial and land surface distribution of fires that produce sufficient smoke to impact different regions of the country. We examine the relative contribution of local versus long-range transport to the presence of smoke in a region as well as the significance of agricultural burning versus wildfires.
2.069 Including the biogeochemical impacts of deforestation increases projected warming of climate.

Early Career Scientist

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Abstract:

Forests cover almost one third of the Earth’s land area and their distribution is changing as a result of human activities. The presence, and removal, of forests affects the climate in many ways, with the net climate impact of deforestation dependent upon the relative strength of these effects (Betts, 2000; Bala et al., 2007; Davin and de Noblet-Ducoudré, 2010).

In addition to affecting surface albedo and exchanging carbon dioxide (CO₂) and moisture with the atmosphere, vegetation emits biogenic volatile organic compounds (BVOCs), altering the formation of short lived climate forcers (SLCF) including aerosol, ozone and methane.

In this work, we combine a land-surface model with a chemical transport model, a global aerosol model, and a radiative transfer model to compare several radiative impacts of
idealised deforestation scenarios in the present day.

Here, we calculate that a scenario of complete global deforestation results in a radiative forcing (RF) of $1.37 \text{ W m}^{-2}$ due to changes in well-mixed greenhouse gases, surface albedo, ozone and the direct and indirect effects of aerosol. We estimate that SLCFs contribute a RF of $0.11 \text{ W m}^{-2}$, 8% the net RF due to deforestation, with the positive aerosol direct ($0.14 \text{ W m}^{-2}$) and indirect ($0.19 \text{ W m}^{-2}$) effects partially offset by the negative RF from changes to ozone ($-0.15 \text{ W m}^{-2}$) and methane ($-0.08 \text{ W m}^{-2}$). We estimate that the positive RF from SLCFs increases the net warming impact from global deforestation by approximately 0.05 K, suggesting that deforestation has a stronger warming impact on climate than previously thought.

References:
Bala, G. et al., 2007, PNAS, 104, 6550-6555.
2.075 Assessment of Emission from Crop Residue Open Burning in Southeast Asia.

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Abstract:
Disposal of agricultural crop residue by open burning (CROB) is increasingly practiced in Southeast Asia (SEA) countries to get rid of huge amounts of this waste annually. The activity generates large amounts of air pollutants which cause deterioration of local air quality and potentially affect the regional climate. This paper reports the emission inventory (EI) results of CROB in 11 SEA countries in 2014. The EI covers major crops in SEA including rice, maize, soybean, potato/sweet potato, groundnut, sugarcane and cassava. The inventory species included trace gases, particulate matter, as well as semi-volatile organic compounds of polycyclic aromatic hydrocarbons (PAHs), dioxins and furans (PCDDs/PCDFs), and greenhouse gases (GHGs). Activity data on field open burning in SEA was primarily collected from the local surveys. The emission factors used in the EI were those developed from the field measurements in the region whenever available. The total annual emissions from SEA in 2014, in Gg, were: 52 \( \text{SO}_2 \); 379 \( \text{NO}_x \); 22,473 \( \text{CO} \); 1,109 \( \text{NMVOC} \); 618 \( \text{NH}_3 \); 1,407 \( \text{PM}_{10} \); 1,288 \( \text{PM}_{2.5} \); 89 \( \text{BC} \); 470 \( \text{OC} \); 209,640 \( \text{CO}_2 \); 640 \( \text{CH}_4 \) and 17 \( \text{N}_2\text{O} \). For toxic air pollutants, the emissions of total PAHs were estimated at 39 Gg while that of PCDD/PCDF was 83 g Toxicity EQuivalent (TEQ). Emission of benzo(a)pyrene (BaP), known as a carcinogenic pollutant, was estimated at 2 Gg in 2014. Indonesia was the top emitter of most species (22-52% depending on species) followed by Vietnam (13-26%) and Thailand (13-19%). The gridded EI results (0.1 degree resolution) prepared in this study can be readily used for regional modeling efforts.
Modulation of nitrogen deposition by natural and anthropogenic land surface heterogeneities.

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Abstract:

Anthropogenic activities have increased N deposition to terrestrial ecosystems by more than threefold relative to preindustrial levels. Such increase has been associated with a wide array of ecosystem responses ranging from increased productivity to eutrophication, soil acidification, and loss of biodiversity. However, significant challenges remain in quantifying the impacts of N deposition on ecosystems in part due to uncertainties in the surface removal of nitrogen, estimates of which are largely based on coarse-resolution (~100km) models. Here, we use the GFDL land-model (LM3) coupled to the GFDL atmospheric chemical model (AM3) to characterize the impact of physical, hydrological, and ecological properties of the land surface on the removal of trace gases and aerosols. In particular, we focus on the sensitivity of reactive nitrogen dry deposition to both natural (e.g., species type, wetness) and man-made (e.g., deforestation, cropping) heterogeneities in the surface properties. We show that land use associated with agriculture increases nitrogen deposition to natural ecosystems both near source regions and in downwind receptor regions. In particular, model estimates of nitrogen deposition to forested ecosystems (in particular coniferous forests) need to be revised upward in regions with significant land-use change. We also show that the response of N deposition to projected changes in nitrogen emissions can vary considerably by land type. In particular, we find that N deposition to North American forests will decline thanks to lower oxidized nitrogen emissions, while N deposition to water bodies will increase in response to higher ammonia emissions. The implications of the heterogeneity in the surface removal of reactive nitrogen for particulate nitrate will also be discussed.

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Abstract:

We describe the measurement strategy and preliminary findings from the Holistic Interactions of Shallow Clouds, Aerosols, and Land-Ecosystems (HI-SCALE) campaign that will be conducted in May and September of 2016 in the vicinity of the DOE’s Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) site located in Oklahoma. Current shallow and deep convective cloud parameterizations used by regional and global models contain uncertainties resulting from insufficient coincident data that couples cloud macrophysical and microphysical properties to inhomogeneity in land use and ecosystems, boundary layer turbulence, and aerosol properties. Rather than targeting specific processes as has been done in previous campaigns, the goal of the HI-SCALE campaign is to provide a detailed set of aircraft and surface measurements needed to obtain a more complete understanding and improved parameterizations of the lifecycle of shallow clouds. Aerosol and aerosol precursor measurements on the research aircraft are made using a High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS), a single particle mass spectrometer (miniSPLAT), and a chemical ionization mass spectrometer (CIMS). Similar measurements will be continuously collected at the surface. The sampling will be done in two periods, one in the spring and the other in the late summer to take advantage of variations in the “greenness” for various types of vegetation, new particle formation, anthropogenic enhancement of biogenic secondary organic aerosol (SOA), and other aerosol properties. The aircraft measurements will be
coupled with extensive routine ARM SGP measurements as well as Large Eddy Simulation (LES), cloud resolving, and cloud-system resolving models. Through these integrated analyses and modeling studies, the affects of inhomogeneity in land use, vegetation, soil moisture, convective eddies, and aerosol properties on the evolution of shallow clouds will be determined, including the feedbacks of cloud radiative effects.
Uncertainty in biogenic emissions from Eucalypts: Implications in urban Southeast Australia.

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Abstract:

Biogenic emissions including isoprene and monoterpenes are strong precursors for ozone and particle formation. In Australian cities, such as Sydney, surrounded by Eucalypt forests, biogenic emissions dominate in summer, leading to deleterious air quality and health implications. Huge distances between Australian cities means that anthropogenic NOx sources are isolated, thus chemistry on the biogenic/urban fringes is of major importance.

Isoprene and monoterpenene measurements are presented from four field campaigns in Southeast Australia; two from Western Sydney (urban), one from Wollongong (coastal urban), and one from Tumbarumba (Eucalypt forest) (Emmerson et al. 2016). The observed concentrations of isoprene and monoterpenes were of a broadly similar magnitude, indicating that Southeast Australia may hold an unusual position where neither chemical species dominates.

The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGANv2.1) was coupled to the CSIRO chemical transport model. The high emission factors within MEGANv2.1 for Southeast Australia over-predicted the observed isoprene (up to a factor of 6) and under-predicted the observed monoterpane concentrations (up to a factor of 4). There was not a consistent factor for either isoprene or monoterpenes that could match observations and modelled results across the four sites/seasons studied.

Re-mapping the Southeast Australian biogenic emission factors is important. A proposed aircraft campaign “Characterising Organics and Aerosol Loading of Australia” (COALA) is aimed for the Austral summer of 2019. COALA will enable study of how these biogenic emissions interact with isolated city plumes. Low NOx Australia may represent future conditions in the Northern Hemisphere, where NOx levels are decreasing due to strengthened air quality regulations.

Abstract:

Biomass burning has the potential to alter numerous land and atmospheric processes, which has strong implications for air quality and feedbacks to the climate system. If plume injection height is incorrect, transport models of those emissions will likewise be incorrect, adversely affecting our ability to analyze and predict climate feedbacks (i.e. black carbon snow/ice, patterns of precipitation, cloud-radiation relationships) and provide timely public health warnings (air quality forecast).

Historically, plume height was based on the pioneering work of G.A. Briggs [1969; 1971] and verified with limited field campaigns. However, presently, there are two instruments that are capable of determining plume injection height, Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) and Multi-angle Imaging Spectroradiometer (MISR). In this presentation, we will demonstrate a novel method that has been developed to estimate biomass burning plume injection height using CALIOP aerosol data, Moderate Resolution Imaging Spectroradiometer (MODIS) Thermal Anomaly data, and the NASA Langley Trajectory Model (LaTM). CALIOP aerosol data are used to initialize aerosol-filled air parcels in the LaTM, which is then run backwards until there is a coincidence between MODIS fire detections and smoke parcels. Specific examples will include smoke transport from North America to the Greenland Ice Sheet and the Tripod fire (July-August 2006), one of the largest fires in the lower 48 in recent U.S. history. Together, CALIOP and MISR can produce the statistical knowledge necessary to improve our understanding of the dynamics of fire plume injection height, thus improving our ability to forecast poor air quality and to accurately define smoke feedbacks with the climate system.
2.082 Accuracy evaluation of MODIS MCD64A1 burned area data product in boreal Eurasia.

Early Career Scientist

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Abstract:

Wildfires play an important role in affecting vegetation dynamics, biogeochemical cycles of carbon, nitrogen and other elements, atmospheric chemistry and the climate. Boreal Eurasia is one of the most important regions where large areas are burned every year. Numerous particles and greenhouse gases are emitted from these fires. These pollutants could be transported to the industrialized region in East Asia, the western North Pacific and the Arctic. Atmospheric transport of fire-emitted pollutants to the Arctic and the consequential deposition is believed to accelerate the arctic warming. To better understand the effect of fires in boreal Eurasia on the arctic, an accurate assessment of burned area from the boreal Eurasia is necessary.

The MCD64A1 burned area product is widely used for global burned area mapping. However, uncertainties in burned area estimations could be introduced due to the “moderate resolution” character of MCD64A1. Therefore, a comparison of MCD64A1 with higher resolution satellite products could provide basic and crucial information for its accuracy assessment and further applications. In this work, we used Landsat 7 surface reflectance, along with a few commercial satellite products as the reference scenes to derive the burned areas in a burning season in 2012. A wide range of ecotypes spanning from the western Russia/Kazakhstan to the eastern Siberia were covered. Each of these burned areas was compared with the corresponding region of MCD64A1 for the same periods. Our preliminary results indicated that MCD64A1 could well capture the large fires, while those less than 100 ha are prone to be undetected. We also found that MCD64A1 tends to underestimate the burned area in general. Based on the error statistics, we suggested the accuracy levels and precautions for applications in each ecotype.
2.083 Global modeling study of dust source and soluble Fe input to the Southern Ocean.

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Abstract:

Atmospheric deposition of dust source materials is a significant source of exogenous iron (Fe) in marine ecosystems. Especially, the Southern Ocean is the most biogeochemically important ocean because of its large spatial extent and its considerable influence on the global carbon cycle. The major sources of atmospheric Fe to the Southern Ocean include South America (Patagonia), Australia, and southern Africa. However, there is large uncertainty in our estimate of the dust emissions in the Southern Hemisphere. Recently, the dust sources in these regions have received considerable attention, both for their potential impacts on climate and ecosystems and for their susceptibility to climate and land-use changes.

Here, we use an atmospheric chemistry transport model to investigate the supply of soluble Fe from dust source regions to the oceans in the Southern Hemisphere. We do so using a physically-based dust emission parameterization to better capture emissions from dry soils in non-vegetated areas. We validated the simulated aerosol optical depth (AOD) using ground-based sun photometer (AERONET) measurements near dust source regions. Our model results suggest that dust emissions from open shrublands in arid and semi-arid regions can be a key contributor to Southern Hemispheric dust, and thus to bioavailable Fe deposition to the Southern Ocean. These results highlight the necessity of improving the process-based quantitative understanding of the dust source fluxes in sparsely vegetated areas with changing climate and land-use practice.
2.084 Secondary Organic Aerosol Production from Herbivore-Induced Plant Volatiles.

Early Career Scientist

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Abstract:

Plant emissions are a major source of biogenic volatile organic compounds (BVOCs) that lead to secondary organic aerosol (SOA) formation. Climate change impacts on BVOC emissions affect the SOA formation potential of the plant emissions, either increasing or decreasing SOA yields depending on the types of emissions that are induced or suppressed. One plant stressor known to have a significant impact on plant emissions is herbivore outbreaks. Herbivore outbreaks are expected to increase as a result of climate change, especially in boreal regions, and can contribute significantly to atmospheric aerosol mass loading. This study investigated the differences in the SOA formation of boreal forest conifer emissions under baseline and herbivore-treatment scenarios in the laboratory.

In this study, BVOC emissions from boreal forest conifers were fed into a flow tube reactor
where they were photochemically oxidized to generate SOA. Emissions from undamaged and damaged saplings were used to generate SOA and results were compared. The herbivore treatment was application of the pine weevil, *Hylobius Abietis*, a bark borer. Plant emissions were monitored continuously with a PTR-ToF-MS. More detailed chemical analysis was performed using cartridge sampling and subsequent analysis with TD-GC-MS. SOA microphysical and chemical properties were monitored with SMPS and HR-ToF-AMS. Results demonstrated that bark borers increase Scots pine monoterpene and sesquiterpene emissions by 75- to 200-fold. Furthermore, the SOA mass yields from the plant emissions were highly variable from experiment to experiment, but could be explained by the different types of monoterpenes the plant was emitting (the monoterpene emission profile). Chemically-speciated terpenoid emission rates from baseline and herbivore-treated plants will be presented along with descriptions of their photochemistry and SOA-forming potentials. These results highlight that herbivore outbreaks could significantly increase SOA production in boreal forest regions in the future.
2.085 Biomass burning emission inventories over Africa: AMMABB and GFED uncertainties investigations.

Early Career Scientist

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Abstract:

Fires occur frequently in most vegetated ecosystems across the world, mostly in tropical and subtropical savannahs where fires are deliberately set during the dry season for multiple purposes. Gases and particles from biomass burning are released into the atmosphere and interact with climate at both local and regional scales. Biomass burning therefore constitutes a complex phenomenon which comprises feedback interactions between ecosystems, humans and climate. Over the past 20 years, several studies have estimated biomass burning aerosol emissions both at global and regional scales. However, large uncertainties still exist in these biomass burning emission inventories particularly over Africa. This study aims to investigate vegetation parameters such as biomass density (BD) and burning efficiency (BE) role in the biomass burning emissions inventories uncertainties. To achieve this purpose, a new inventory over Africa was developed for 2001-2012 using MODIS fire product, Global Land Cover (GLC) vegetation map, current emission factors and same ‘bottom-up’ methodology were used. Moreover, sensitivity tests on factors of African Monsoon Multidisciplinary Analysis Biomass Burning (AMMABB) and Global Fire Emission Database (GFED) vegetations parameters were conducted. Results showed that, BD and BE are important parameters for biomass burning emissions in Africa and explained the large differences observed between AMMABB and GFED emissions inventories.
Measurement of isoprene emissions from UK woodland using an airborne platform.

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Abstract:
Ozone (O₃) is an oxidising agent that is both harmful to human health and damaging to crops and natural vegetation. It is produced by the photochemical oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides (NOₓ). Although precursor emissions have decreased over time in the UK, these reductions have not uniformly resulted in the anticipated decreases in background and peak O₃ concentrations, leaving critical questions about the relative importance of the various sources. Biogenic VOC emissions are one area of particular uncertainty. Canopy or landscape scale emission rates may be obtained by extrapolated leaf-level or whole plant measurements of emission rates, accounting for biomass, light and temperature distributions, however, since there are both hugely varying emission factors for different plant species and highly complicated “activity factors” driven by light, temperature, soil moisture, biomass etc., “bottom up” emission inventories for biogenic VOCs have very great uncertainties associated with them.

Airborne flux measurements offer the possibility of obtaining large scale emission estimates, but at the spatial and temporal resolution necessary for understanding ground-level pollutant formation and exposure. Measurements of isoprene fluxes using virtual disjunct eddy covariance were made in summer 2013 and 2014 over rural areas of southern United Kingdom from a low flying aircraft. Fluxes of up to 2 mg m⁻² hr⁻¹ were measured, with the highest measurements observed over areas dominated by broadleaf woodland. Measured isoprene emissions were compared to those used in the EMEP4UK regional atmospheric chemistry transport model and whilst good agreement was seen in places, the model showed a significant underestimation in the areas of the highest measured emissions. The potential effect of this underestimation on the prediction of O₃ in the EMEP4UK model was also investigated.
2.088 Comparison of biomass burning inventories processed by GEOS-Chem with total column and satellite data in Australia.

Early Career Scientist

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Abstract:

Australia contributes approximately 6% of global biomass burning CO\textsubscript{2} emissions, mostly from savanna type fires. This estimate comes from biomass burning inventories that use emission factors derived from field campaigns performed outside Australia. The relevance of these emission factors to the Australian environment has not previously been evaluated and therefore needs to be tested. Here we compare predictions from the chemical transport model GEOS-Chem and the global chemistry-climate model ACCESS-UKCA run using different biomass burning inventories to total column measurements of CO, C\textsubscript{2}H\textsubscript{6} and HCHO, in order to identify the most representative inventory for Australian fire emissions. The measurements come from the Network for Detection of Atmospheric Composition Change (NDACC) and Total Carbon Column Observing Network (TCCON) solar remote sensing Fourier transform spectrometers and satellite measurements from IASI and OMI over Australia. We evaluate three inventories: the Global Fire Emission Database version 4 – GFED4 (Giglio et al. 2013), the Fire Inventory from NCAR – FINN (Wiedinmyer et al. 2011), the Quick Fire Emission Database – QFED from NASA and the MACCity emission inventory (from the MACC/CityZEN EU projects; Angiola et al. 2010). From this evaluation we aim to give recommendations for the most appropriate inventory to use for different Australian environments. We also plan to examine any significant concentration variations arising from the differences between the two model setups.
2.089 Investigating the effects of aged wildfire smoke on photochemistry in the Northern Front Range of Colorado.

Early Career Scientist

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Abstract:

Wildfires are likely to increase in frequency and intensity in the future and human populations are likely to be increasingly affected by wildfire smoke. Wildfire smoke contains a complex mixture of trace species, including aerosols and ozone precursors. The Northern Front Range Metropolitan Area of Colorado (encompassing Denver and the surrounding municipalities along the eastern edge of the Rocky Mountains) is one of the fastest growing metropolitan regions in the country and new technologies have led to a boom in oil and gas extraction in the adjacent Denver-Julesberg Basin. Here we present an analysis of two aged wildfire plume events in the Front Range based on data collected at the NOAA Boulder Atmospheric Observatory (BAO) tower in northeastern Colorado during summer (3 July through 7 September) 2015. During this campaign, we measured a broad suite of VOCs, NO\textsubscript{x}, and several oxidized nitrogen species (nitric acid, peroxy acyl nitrates (PANs), and alkyl nitrates) in addition to O\textsubscript{3}, CH\textsubscript{4}, CO\textsubscript{2}, and CO. Each smoke event lasted several days, and included weekend and weekday days. From 6 July through 10 July smoke from wildfires in British Columbia was present in the Front Range and 16 August through 26 August was influenced by smoke originating in Washington and Oregon. The presence of smoke is apparent in the data as strongly elevated CO (a mean of 259 ppbv during smoke events vs. 161 ppbv during rest of campaign), PAN, PPN and benzene. Smoke was present on 6 out of 21 high ozone days during the campaign (defined as hourly O\textsubscript{3} > 70 ppbv).
2.092 Fungal content in aerosols at the Caribbean region during African Dust incursions.

Early Career Scientist

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Abstract:

African Dust (AD) particles impacted the Caribbean region every year during the summer months causing an increase in particulate matter concentrations. Recent studies suggest that AD incursions have the ability to transport particles of biological origin through long distances. Fungal spores are a major component of primary biogenic aerosol particles, are ubiquitous in the atmosphere, and play an important role in the chemistry and physics of the atmosphere, climate, and public health. The relationship between AD incursions and the concentration of fungal spores in the Caribbean region is poorly understood. Preliminary results using a Burkard spore trap to determine spore’s concentration at the tropical montane cloud forest of Pico del Este (PE) at El Yunque National Forest in Puerto Rico (PR), showed a considerably decrease (by as low as 75%) in the concentration of fungal spores during AD events. To have a better understanding of the effects of AD incursions on fungal spore’s emissions, aerosol samples in the presence and absence of AD were collected using the stacked-filter units at the University of Puerto Rico’s (UPR) atmospheric observatory of Cabezas de San Juan (CSJ), a marine site located in Fajardo, PR; and at an urban site, Facundo Bueso building, at the UPR. The use of satellite images of aerosol optical thickness, and the results from air masses backward trajectories calculated with the NOAA HYSPLIT model were used to determine the presence of AD incursions during the sampling periods. At the meeting we will present preliminary results on gravimetric analyses, ion speciation, and carbonaceous content (organic and elemental carbon (OC and EC)) together with culture-independent molecular analysis, i.e., amplicon sequencing of the ITS and LSU rRNA genes, to assess which fungal taxa are present and their dynamics in AD vs. non-AD samples.
Abstract:

Dimethyl sulfide (DMS) plays a pivotal role in the climate change-related biogeochemical cycles. Because the oxidation of DMS contribute to the formation and growth of aerosol, it is an important precursor of cloud condensation nuclei (CCN), and thus essential to understand the change of global radiative forcing budget. The oceanic emission is a dominant natural source of atmospheric DMS. The oceanic DMS is produced by biological processes while its production rate is widely varied by the ocean’s biological environmental characteristics.

In the polar region, the oceanic DMS is essential to understand and predict climate variability. In the Southern Ocean, DMS has been significantly underestimated. Also, high productivity of Antarctic coastal polynyas implies strong DMS emissions from the Antarctic polynyas. In the Arctic Ocean, rapid sea ice decline will clearly impact ocean environment including biological productivity, and thus alter DMS emission strength as well. Therefore, monitoring DMS in the polar oceans are essential to understand climate change processes. However, its observations are significantly lacked due to the limited accessibility of polar oceans.

Membrane inlet mass spectrometry (MIMS) technique directly samples analyte gases from the aqueous phase gases in seawater through a semi-permeable membrane. Since this method does not require headspace equilibration, MIMS enables us to make a near-real time continuous observation of dissolved gases. Thus, it is especially useful to capture environmental changes in oceanic waters showing a significant concentration variation or occurring an active physical, chemical and biological processes.

We have made continuous high frequency DMS observations in the Southern Ocean (Jan. – Feb. 2016) and the Arctic Ocean (Aug. 2016) using MIMS on the Korean icebreaker R/V Araon. Here, we will present the results of oceanic DMS measurements from the two research cruises as a beginning of our effort to understand climate change and polar ocean environment change.
2.094 VOC observations over the Southern Ocean during ORCAS.

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Abstract:

The 2016 O₂/N₂ Ratio and CO₂ Airborne Southern Ocean Study (ORCAS) provided a unique opportunity to investigate the behaviors of short-lived VOCs over the Southern Ocean during the biologically-productive summer months in the context of the global carbon cycle. The primary goal of the study was to investigate the air-sea exchange of O₂ and CO₂ over the Southern Ocean where the uptake of anthropogenic carbon is a key process that is poorly represented by models. Measurements of halocarbons, DMS, OVOCs and anthropogenic trace gases were made using the Trace Organic Gas Analyzer (TOGA) and the Advanced Whole Air Sampler (AWAS) alongside measurements of O₂ and CO₂ and other gases from the NSF/NCAR GV aircraft. The ORCAS research flights, conducted between southern South America and the Antarctic Peninsula, included multiple dips into the boundary layer and extended to 12 km in altitude. Preliminary results from the study indicate strong positive correlations between O₂ and several observed halogenated VOCs in the marine boundary layer and lower free troposphere, and inverse correlations between O₂ and acetone, HCN and CH₃CN. In addition, VOC observations can be used to investigate the transport of anthropogenic and continental trace gases from lower latitudes to the very pristine Southern Ocean atmosphere. We will explore our observations in the context of temporal and spatial variability, and highlight some of the key findings from the field campaign.
2.095 Monoterpene chemical speciation at Amazonian Tall Tower Observatory (ATTO) tropical rainforest.

Early Career Scientist

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Abstract:

Little is known about biological and chemical processes happening within the tropical rainforest canopy. Especially, chemical speciation of monoterpenes in the Amazonian atmosphere is rare but important in order to understand the overall reactivity of the monoterpenes emitted from the forests as well as to understand why plants emit diverse mixtures of monoterpene species at different rates. The objective of this study was to analyse the chemical speciation of monoterpenes within the canopy at the tropical rainforest of the Amazonian Tall Tower Observatory (ATTO). The chemical identification was performed by Gas Chromatography Flame Ionization Detection (GC-FID). For sampling two autosamplers positioned at a tower at 12 and 24 m were used. Sampling of volatile organics occurred on cartridges filled with carbograph adsorbents. Samples were collected during October 2015, and were compared with previous studies at the site. Alternatively, vertical profile measurements (0.05, 0.5, 4, 12, 24, 38, 53 and 79 m) of total monoterpene measurements determined by Proton-Transfer Reaction Mass Spectrometry (PTR-MS). The results showed a distinct chemical speciation along the heights. For instance, b-pinene and a-terpinene are more abundantly found at 12 m, suggesting differences in reactivity within the canopy. The most abundant monoterpenes at both heights are a-pinene, limonene and myrcene. We discuss if such differences are due to emission differences due to different plant species, a difference in leaf developmental stage, or other factors, such as phytopathogenic infections or ozone regimes that prove this difference in the speciation.
2.096 New understanding on sources and impacts of marine VOCs from the Oceanic Reactive Carbon: chemistry-climate impacts (ORC3) project.

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Abstract:

Oceanic emissions of reactive organic carbon alter marine background chemistry and have the potential to contribute to marine aerosol distributions, with implications for climate. Highly reactive compounds such as isoprene, monoterpenes and glyoxal have been observed in the marine atmosphere, yet their atmospheric impact is extremely uncertain. The Oceanic Reactive Carbon: chemistry-climate impacts (ORC3) project aims to improve our knowledge of the sources and impacts of these reactive species in the remote marine atmosphere. The project included two 1-month field campaigns at the Cape Verde observatory in the tropical Atlantic Ocean. New datasets on the abundances of speciated monoterpenes, isoprene and glyoxal were obtained, spanning several weeks. Glyoxal was measured using a sensitive laser-induced phosphorescence (LIP) in-situ
technique. Cruise observations of isoprene and monoterpenes have also been made in air
and seawater during cruises traversing the north and south Atlantic, and in the Arctic
Ocean. Both box modelling and global modelling studies exploiting these new datasets
have been used to investigate sources, processing and impacts of these reactive organic
compounds.

Maximum glyoxal mixing ratios of ~15 pptv were observed during ORC3, with typical
daytime maxima of ~ 5-6 pptv during June and ~ 6-8 pptv during August-September.
Using Master Chemical Mechanism (MCM) box model simulations constrained to
observations of glyoxal precursors, we investigate the sources and sinks of glyoxal in the
tropical marine boundary layer. Total monoterpane observed at Cape Verde and during
the cruises was present in very small concentrations (1-4 pptv), however several different
terpenes were observed. Using a global model, we have constrained a biologically-driven
source term for monoterpenes to these observed atmospheric concentrations, and
derive a source of ~5-10 Tg/yr from the global oceans. Using a global aerosol-chemistry
model, we investigate the implications of this terpene source for remote marine aerosol
and cloud condensation nuclei abundances.
2.098 Impacts on Ozone of a New Interactive Soil NOx Scheme.

Early Career Scientist

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Abstract:

Ozone chemistry in remote regions of the troposphere is often NOx-limited, and soil NOx emissions therefore make an important contribution to ozone production in these areas. Atmospheric chemistry models generally use prescribed monthly mean soil NOx emissions. Since real-world soil NOx emissions are highly sensitive to temperature and rainfall, a soil NOx scheme that includes these factors will result in improved temporal resolution for not only NOx emissions but also for ozone concentrations and the oxidative capacity of the atmosphere. Soil NOx is produced through nitrification, denitrification, and chemodenitrification. These processes depend on soil temperature, pH, moisture, and nitrogen content. Comprehensive schemes for soil NOx production should therefore include these factors, as well as vegetation type, as a substantial proportion of emitted soil NOx is taken up by the canopy. We are developing a model that predicts soil NOx emissions interactively based on soil moisture, soil temperature, nitrogen availability, pH, and vegetation type. We will present results from this model and explore the impact of our new representation of soil NOx emissions on tropospheric ozone.
2.099 Emission Factors of Trace Gases and Particulates from Australian Savanna Fires.

Early Career Scientist

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Abstract:

In June 2014 a measurement campaign took place at the Australian Tropical Atmospheric Research Station (ATARS) in the Northern Territory, Australia, aimed at characterising the emissions from early dry season savanna fires. The campaign was especially focused on understanding aerosol composition and size distribution. Equipment deployed to measure aerosol properties included a multi-angle absorption photometer, a nephelometer, a cloud condensation nuclei counter, a condensation particle counter, two scanning mobility particle sizer, two aerosol mass spectrometers (one a time of flight instrument), a multi-axis differential optical absorption spectrometer, a volatility-humidity tandem differential mobility analyser and two high volume aerosol samplers (one PM10 and one MOUDI). In addition there were measurements of mercury in both gas and aerosol phase. Complementary measurements of trace gases were provided by a proton transfer reaction mass spectrometer, a volatile organic compounds sequencer, a Fourier transform spectrometer, an ozone analyser and a nitrogen oxides monitor. This poster will present results from the Fourier transform spectrometer, the N-nitrogen oxides monitor, the scanning mobility particulate sizer, the Tekran mercury analyser and the aerosol mass spectrometer. In particular individual fire events have been identified and emission factors calculated for CO2, CO, CH4, N2O, NOx, elemental mercury and chemically speciated aerosols, and aerosol size modes (Aitken and Accumulation).
2.100 Chemical Mechanisms in the Forest Canopy: Understanding the HOx-NOx-BVOC Triad.

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Abstract:  
The emissions of biogenic volatile organic compounds (BVOC) are known to exert significant control on tropospheric composition and the oxidation capacity of the atmosphere through (1) the formation of ozone in the presence of reactive nitrogen oxides (NO$_X$ = NO + NO$_2$) and sunlight, (2) the formation of secondary organic aerosols (SOA) through the transformation of its oxidation products, and (3) the regulation of hydrogen oxide radicals (HO$_X$ = OH + HO$_2$). Recently, new pathways of BVOC oxidation have been identified in low-NO$_X$ regions that rectify discrepancies between observations and BVOC-NO$_X$-HO$_X$ chemistry in regional and global transport models. However, these mechanisms do not show consistent improvement across a range of NO$_X$ concentrations. Here, we discuss the use of different chemical mechanisms within the forest canopy to understand atmospheric chemistry across a range of HOx-NOx-BVOC concentrations. We highlight how different types of forest canopies under a range of anthropogenic influence behave in the HOx-NOx-BVOC triad, with the goal of elucidating how lumped chemical mechanisms can be improved to simulate this full parameter space.
3.001 The impacts of solid fuel interventions and electrification on ambient air quality in India.

Early Career Scientist

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Abstract:

Combustion of solid fuels for cooking and heating is widespread across India with large but poorly quantified impacts on ambient air quality. Over the past decade there have been substantial changes in residential energy use across India, due to electrification schemes and implementation of large-scale clean cookstove interventions. We investigate the impacts of changing residential solid fuel combustion over the period 2000 to 2015 on ambient particulate air quality in India. We synthesise information on the location and size of solid fuel interventions, such as the National Biomass Cookstove Initiative. To assess changes in electrification we use data from the National Oceanic and Atmospheric Administration’s (NOAA) National Geophysical Data Centre (NGDC) Version 4 Nighttime Lights Time Series from the Defence Meteorological Satellite Program (DMSP) and the Visible Infrared Imaging Radiometer Suite (VIIRS). We also use electrification data from the International Energy Agency (IEA) and The World Bank Group. We assess the impacts of such changes on air quality using the Weather Research and Forecasting Model with Chemistry (WRF-Chem) regional air quality model, remote aerosol monitoring networks, and satellite observations. Our study will inform the potential for electrification and solid fuel interventions to improve ambient air quality and human health.
3.004 The atmospheric response to building a large-scale space-based solar power system.

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Abstract:

New rocket technology using hydrogen-burning engines may allow a vast number of relatively low cost flights to space in the near future. One such proposal for this new rocket technology is to build a space based solar power system. It is estimated that 10^5 flights per year for 10 years could build a space station capable of replacing about 10% of today’s global electricity production. The emissions from these flights could pose a potential risk to the climate and stratospheric ozone. The effects of the emissions from these rocket engines are quantified for 10^4-10^6 flights per year using the CESM-WACCM climate/chemistry model and the NOCAR two-dimensional model. Water vapor from combustion is the primary emission product, however a substantial amount of NO\textsubscript{X} is produced in the superheated engine exhaust and upon reentry. A total of 10^5 flights per year more than doubles the amount of water in the mesosphere and increases the stratospheric water vapor by roughly 10%. This added water vapor increases high altitude clouds at the poles but its effect on ozone is relatively small. The NO\textsubscript{X} production from spacecraft re-entry is more than an order of magnitude larger than the natural production from meteors and destroys a substantial amount of ozone. At 10^5 flights per year, a reduction by about 1.5 Dobson units or roughly 0.5% occurs in the column integrated global ozone abundance. The largest losses occur in the polar regions. This is a large perturbation, but much less than at the peak of ozone loss in 1998 when global ozone was diminished by 4% due to anthropogenic emissions.
3.006 Top-down estimates of methane and nitrogen oxide emissions from the Haynesville and Fayetteville shale gas production regions.

Early Career Scientist

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Abstract:

Production of unconventional natural gas grows rapidly during the past ten years in the US. This increase most likely led to an increase in emissions of methane (CH$_4$) and nitrogen oxides (NOx). In terms of radiative forcing, CH$_4$ is the second most important greenhouse gas after CO$_2$. NOx is a precursor to ozone (O$_3$) in the troposphere and nitrate particles, both of which are regulated by the US Clean Air Act. However, the emission inventories of CH$_4$ and NOx from the shale regions are highly uncertain. To improve the inventories, we use data collected during the Southeast Nexus of Climate Change and Air Quality (SENEX) aircraft campaign (June-July, 2013) to drive inversion calculations and estimate CH$_4$ and NOx emissions in the Haynesville and Fayetteville shale production regions. We use three transport models and EPA’s 2011 National Emission Inventory as prior information to optimize CH$_4$ and NOx emissions, taking advantage of a Bayesian inversion technique. The posterior CH$_4$ emissions are then used to constrain NOx emission estimates as well using a flux ratio inversion technique. Compared with the ground-based in-situ observations, the optimized CH$_4$ and NOx inventories improve the ground level CH$_4$ and O$_3$ concentrations simulated with the Weather Research and Forecasting model coupled with chemistry (WRF-Chem).
3.007 Emissions of volatile organic compounds (VOCs) from oil and natural gas activities in 13 major U.S. shale basins.

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Abstract:

The recent and unprecedented increase in oil and natural gas production from shale formations is associated with a rise in the production of methane (CH$_4$) and non-methane volatile organic compounds (VOCs) including natural gas plant liquids (e.g., ethane, propane, and butanes) and liquid lease condensate (e.g., pentanes, hexanes, aromatics and cycloalkanes). Methane is an important greenhouse gas and the emission of VOCs may affect local and regional air quality due to the potential to form tropospheric ozone and organic particles as well as from the release of toxic species such as benzene and toluene. Since 2010, the production of natural gas liquids and the amount of natural gas vented/flared has increased by factors of approximately 1.3 and 1.6, respectively (U.S. Energy and Information Administration), indicating an increasingly large potential source of hydrocarbons to the atmosphere.

Here we present measurements of a full suite of carbon-containing gases emitted from oil and gas activities in 13 U.S. shale basins in order to better understand their potential air quality and climate impacts. These measurements were made between 2011-2015 aboard the NOAA WP-3D research aircraft or at various ground sites. Aircraft measurements of non-methane hydrocarbons were measured using a whole air sampler (WAS) with post-flight analysis via gas chromatography-mass spectrometry (GC-MS). The discrete whole air samples are complimented by near-continuous measurements of
methane (Picarro spectrometer), ethane (Aerodyne spectrometer), and VOCs (H$_3$O$^+$ chemical ionization mass spectrometer). We will (i) compare the composition of methane and VOC emissions for each basin in order to show that each shale basin has a unique VOC source signature, and (ii) quantify emission fluxes of VOCs for a subset of these basins. This information is critical for accurate emission inventories and input for air quality models.
**3.008 Mobile Field Measurements of Methane Gas Using Open-Path Cavity-Ring-Down spectroscopy.**

Early Career Scientist

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Abstract:

Methane is a significant atmospheric pollutant and greenhouse gas, approximately ~37x stronger than carbon dioxide, and it is becoming increasingly important to quantify emissions from leaks during extraction, production, processing, transportation and storage of natural gas. Recent work has demonstrated that cavity ring-down spectroscopy (CRDS) can be applied in open-path configurations while still achieving relatively high optical sensitivity allowing for detection of ppb level methane increases. Influence due to ambient aerosols is reduced through the use of software filters which remove fluctuations due to particles while preserving the signal as a result of gasses. Spectral peaks are significantly broadened in atmospheric pressure but can be fit using a synthetic spectrum consisting of a sum of relevant Lorentzian peaks, including interfering species. The high-reflectivity cavity mirrors are kept clean using a simple purge mechanism consisting of a micro-diaphragm pump and hepa filter which pull ambient air into a positive pressure enclosure at the mirror surface. We present work showing controlled release and preliminary field measurements using a compact open-path CRDS instrument operating at 1651 nm. Open-path CRDS decreases the weight and power requirements of the sensor so that it can be powered using a simple battery and mounted on mobile platforms such as an automobile or unmanned aerial vehicle (UAV). Our sensor is deployed on the roof of a small truck with a cable connecting the sensor package to a module containing all electronics, controls and power conditioning. Results of field measurements are discussed, including the influence of ambient aerosols, wind and proximity to leaks.
3.009 Field studies quantifying atmospheric emissions from energy production and consumption in the U.S.

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Abstract:

Fossil-fuel energy production and consumption invariably leads to atmospheric emissions that can affect air quality, climate, and human health. I will summarize recent findings from field studies designed to quantify emissions from specific sources representing major energy-producing and -consuming sectors in the U.S. These findings illustrate spatial and temporal variability of emissions on daily, seasonal, and decadal time scales, and suggest the need for improved quantification and attribution of energy-related emissions that could be provided by a more integrated observing network.

Early Career Scientist

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Abstract:

Atmospheric pollutant emissions have serious consequences for ambient air quality, climate, and public health. Emissions, such as Particulate Matter (PM), Oxides of Nitrogen (NO\(_x\)), Carbon Monoxide (CO) and Sulfur Dioxide (SO\(_2\)), are rapidly changing in China. Residential energy usage is a large contributor to ambient PM\(_{2.5}\), but plans are underway to mitigate these emissions through installation of improved cookstoves in rural China. Significant decreases in SO\(_2\) and NO\(_x\), largely a result of policies controlling thermal power plant emissions and mobile sources, have already been observed over (approximately) the last five years, as has been tracked by remote sensing observations. These rapid changes make it challenging to accurately simulate air quality in China using existing emission inventories. Further, assessment of specific pollution mitigation efforts, using chemical transport models, is limited by inaccurate emissions.

We conducted a series of sensitivity studies to investigate the impacts of rapidly changing emissions on air quality in China. We investigated several factors, such as the changes to NOx and SO\(_2\) emissions based on the latest NASA Ozone Monitoring Instrument (OMI) data estimates, and the sensitivity of ambient pollutant concentrations to current and potential changes in residential emissions, distinguishing estimates of heating emissions from cooking. Annual simulations were completed for 2014 using the Weather Research and Forecasting model with Chemistry (WRF-Chem), a fully-coupled, “online” regional chemical transport model. Model output was evaluated against surface...
air quality measurements across China, with the evaluation results used to assess the accuracy of available emission estimates. Finally, applying model output to available exposure—response relationships for PM$_{2.5}$ and cardiopulmonary health outcomes, we estimate potential health benefits from future emission mitigation scenarios associated with household energy interventions.

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Abstract:

With technological growth and the development of unconventional fossil fuel resources, the volume of oil and gas extracted from basins across North America has
grown dramatically over the past 5-10 years. This has had significant, observable effects on ambient air quality, including enhancements in ozone and secondary organic aerosol (SOA) formation, but the extent of emissions across the volatility range of gas-phase organic compounds remains uncertain. Using offline analysis and a survey of gas-phase organic compound samples from a mix of laboratory, ground site, mobile lab, and aircraft platforms; we examine the magnitude and detailed chemical composition of emissions and ambient concentrations of volatile organic compounds (VOCs), intermediate-volatility organic compounds (IVOCs), semivolatile organic compounds (SVOCs).

Offline samples of compounds with 7 or more carbon atoms are collected on custom adsorbent traps and analyzed using a thermal desorption-gas chromatography system connected to both a very-high resolution, chemical ionization, tandem mass spectrometer and an electron ionization quadrupole mass spectrometer. The sampled complex mixtures of organic compounds are comprehensively speciated by carbon number and compound class, with resolution of major isomers. We present these results in an intercomparison across several North American basins with a varying mix of shale oil and shale gas operations. We also present the results of a laboratory experiment that realistically simulates the processing of oil sands, which has been shown to cause SOA formation downwind of extraction and processing operations in Alberta. We compare the distribution of chemical compound classes and volatility across all measured sources with a preliminary assessment of their relative potential for SOA and ozone formation.
3.018 Characterization of a Quantum Cascade Tunable Infrared Laser Differential Absorption Spectrometer (QC-TILDAS) for atmospheric ethane and methane field measurements.

Early Career Scientist

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Abstract:

Ethane, the second most abundant hydrocarbon in the atmosphere and a tropospheric ozone precursor, has the potential to influence regional air quality. Ethane can also help to inform the understanding of climate forcing, as measurements of ethane are of interest for use as a constraint on methane emissions attribution. The emission ratios of ethane and methane from oil and natural gas operations, a major source of both compounds, can be used to constrain methane emissions in large-scale modeling and for methane source attribution on smaller scales, such as in oil and natural gas basins. Here we characterize a tunable infrared laser differential absorption spectrometer (TILDAS), a mini laser trace gas monitor manufactured by Aerodyne Research, Inc. The quantum cascade laser at 2990 cm$^{-1}$ allows for measurement of both ethane and methane mole fractions, eliminating the need for additional instruments, calibration of multiple instruments, and post-deployment time alignment. The fast response (1 Hz or less) of the instrument permits deployment on various mobile platforms. We present results from tests conducted in the laboratory and aboard ground-based and airborne platforms. Configuration and parameters needed to optimize deployment on these mobile platforms are discussed. Characterization of instrument response time, noise, calibration drift, and potential interferences will be presented. Complete characterization of this instrument will allow for its use for various applications, including for the purpose of long-term
monitoring or in intensive field campaigns.
3.019 Open-Path Hydrocarbon Laser Sensor for Oil and Gas Production Facility and Fenceline Monitoring.

Early Career Scientist

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Abstract:

We are developing a new open-path laser absorption sensor for measurement of unspecified hydrocarbons for oil and gas production facility monitoring. Such measurements are necessary to meet regulations, to quantify greenhouse gas emissions, and to detect volatile organic compounds (VOCs) that may have adverse health effects or act as precursors to ozone formation. The present contribution presents a proof-of-principle demonstration of an open-path laser absorption sensor. Our initial design employs a single path measurement system though future implementations may use multiple paths for large scale facility monitoring. For example, a laser at a central location could be directed to multiple retro-reflectors around a perimeter or could target equipment and areas of interest given specific operational conditions. The laser sensor uses a compact mid-infrared (MIR) interband cascade laser at ~3.41 µm to measure absorption of several contributing hydrocarbon species over open-paths of ~50-100 m. Spectral simulations show that for typical concentrations of interest, the laser transmission drops by greater than ~10% providing a robust measurement. The contribution will include: design and laboratory (closed-path) testing of the sensor and initial outdoor open-path field tests. We will discuss signal levels, sensitivity, and signal fluctuations due to turbulence (beam steering) and aerosol extinction over the long path.
3.020 Coal seam gas and air quality in the Surat Basin, Australia: monitoring and modelling the impacts.

Early Career Scientist

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Abstract:

Coal seam gas (CSG) production has expanded rapidly in the state of Queensland, Australia in the past decade, driven by three major CSG to liquefied natural gas (LNG) export projects. The Surat Basin, a predominantly agricultural region with vast gas reserves, is the hub of this development.

A new ambient air quality study has been instigated in the Surat Basin as part of the Gas Industry Social and Environmental Research Alliance (GISERA), a partnership between the federal government’s Commonwealth Scientific and Industrial Research Organization (CSIRO) and industry partners. This is the first comprehensive air quality study to be undertaken in an unconventional gas region of Australia. The study involves monitoring NO\textsubscript{X}, O\textsubscript{3}, total VOCs, CO, CH\textsubscript{4}, CO\textsubscript{2}, PM2.5 and PM10 at three ambient air quality sites in the heart of the gas fields, and NO\textsubscript{X}, O\textsubscript{3}, CO, CH\textsubscript{4}, CO\textsubscript{2} at sites upwind and downwind. Ambient air quality data will be live streamed to the Queensland Government air quality site, ensuring maximum transparency for concerned communities. Additional measurements include ambient passive speciated VOC, aldehyde and H\textsubscript{2}S measurements, and detailed characterization of potential CSG related emission sources, including fugitive gas, gas fired compressors and engines, feed pond water and naturally occurring radioactive materials.

A CSIRO meteorological model coupled with a chemical transport model is being used to examine the distribution of primary pollutants (including NO\textsubscript{X}, CO) in the Surat Basin as well as secondary pollutants (including O\textsubscript{3} and secondary PM). The study will explore the contribution from the CSG industry and other sources to observed levels of pollutants.
over one year at 1km, hourly resolution.
We provide an overview of the study and some preliminary ambient air monitoring data,
gas composition data, and air quality modelling results. Future unconventional gas
development in Australia will be discussed, with potential implications for air quality.
3.021 First Emission Estimates From Oil and Gas Exploration and Production Activities in the Norwegian Sea.

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Abstract:
The Arctic warms twice as fast as the rest of the world, and the Arctic Ocean is projected to become nearly ice-free in summer within this century. This opens the possibilities for hydrocarbon extraction, which will result in an increase of local emissions of short-lived climate forcers and greenhouse gases in a very sensitive region. Type and quantity of emissions of off-shore oil/gas exploration are not well known and very few independent measurements exist. During the ACCESS (Arctic Climate Change, Economy and Society aircraft) campaign in 2012 and one follow-on mission in 2016, the DLR Falcon conducted measurements in the Norwegian Sea area. The objective of these missions were to study chemical composition of emissions and to quantify source strengths of climate forcers and precursors (NO$_x$, SO$_2$, CO$_2$, CH$_4$ and non-vol. particles) released by oil/gas production. The measurements, carried out in cooperation with the Statoil company, focused on the largest platforms in that area, including oil and gas production facilities, drilling rigs and storage tankers. Elevated levels of most trace gas and aerosol concentrations were observed downstream of each facility. Using high resolution particle dispersion modelling, the measurements are used to constrain atmospheric emissions of off-shore oil/gas production in the Norwegian Sea.
3.022 Black carbon emissions from transport activity in Chile: historic trends and 2050 forecast.

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Abstract:

Historic fuel consumption trends from transportation modes in Chile are provided by different organizations, applying diverse methodologies and using multiple time scales, making difficult a comprehensive energy assessment of the sector for the whole country. This research provides results of a 4-year study aiming at building a database of historic and future emission and energy trends from transportation modes in Chile (land, air and sea), region by region, from 1998 until 2050. Specifically for on-road vehicles, black carbon emissions are inferred from fine particle matter emissions from different vehicle categories and engine types, using international correlation factors adjusted for local conditions in Chile. Future projections show a trend of reduction in black carbon emissions between 2020-2038, followed by a slight increase during 2040-2050. This behaviour could be explained by the interaction between the introduction of new technologies (particle filters, electric vehicles) versus a permanent growth of activity in terms of vehicle kilometers travelled.
Assessment of Satellite Capabilities to Detect Impacts of Gas and Natural Oil Activity, from Analysis of SONGNEX 2015 Aircraft Measurements.

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Abstract:

In the last decade, the rate of domestic energy production from oil and natural gas has grown dramatically, resulting in increased concurrent emissions of methane and other volatile organic compounds (VOCs). Products of VOC oxidation and radical cycling, such as tropospheric ozone (O$_3$) and secondary organic aerosols (SOA), have detrimental impacts on human health and climate. The ability to monitor these emissions and their impact on atmospheric composition from remote-sensing platforms will benefit public health by improving air quality forecasts and identifying localized drivers of tropospheric pollution.

Instruments to be deployed on upcoming satellites, such as TROPOMI (2016 launch) and TEMPO (2018/19 projected launch), will be capable of measuring chemical species related to energy drilling and production on unprecedented spatial and temporal scales, however there is need for improved assessments of their capabilities with respect to specific applications. We use chemical and physical parameters measured via aircraft in the boundary layer and free troposphere during the Shale Oil and Natural Gas Nexus (SONGNEX 2015) field campaign to view chemical enhancements over tight oil and shale gas basins from a satellite perspective. Our in-situ data are used to calculate the
planetary boundary layer contributions to the column densities for formaldehyde, glyoxal, O$_3$, and NO$_2$; these emulated measurements are compared to TEMPO and TROPOMI capabilities in order to assess the degree to which their retrievals will be able to discern the signatures of oil and natural gas activity, and to ascertain the information that may be derived from trace gas enhancements.
3.030 Emissions of NOx, SOx, and PM from stationary combustion sources in Brazil: Spatial distribution and population exposure assessment.

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Abstract:

The emissions and the spatial distribution of the major stationary sources of NOx, SOx, and PM in Brazil were estimated in this work. A total of 16 refineries, 96 cement industries, 64 pulp and paper industries and 1730 thermo power plants (natural gas, sugarcane bagasse, residual fuel oil, diesel oil and coal) were included in the inventory. The emissions were calculated based on emission factors (EF) within a range varying from lower to higher limits proposed by the AP-42 standards of the US Environmental Protection Agency. The estimates were 857±415 Gg/year for NOx, 1,51±1,23 Tg/year for SOx, and 10,4±10,1 Tg/year for PM. Refineries, cement industries and diesel power plants are the dominating sectors for NOx emissions, with 28% (26), 39% (24), and 21% (36), respectively, considering lower(higher) EF. Refineries dominate the emission of SOx (42%), followed by diesel (18%), fuel oil (18%), Coal 15%, and Cement (8%) under lower
EF. At higher EF refineries emitted 44%, followed by coal power plants (33%) and cement industries (15%). In terms of PM, under lower EF limits, coal power plants represent 65% of emissions, followed by refineries (21%). On the other hand, at higher EF limits, the cement industry sector dominates, with 78%, followed by coal power plants with only 11%. Comparing the emissions by stationary sources, estimated by intermediate values of EF, with the vehicular emissions, estimated by the Brazilian Ministry of the Environment (MMA), emissions of NOx are slightly lower than vehicular, while the emissions of SOx and PM are 300 times higher than vehicular. The contribution of stationary sources are still significant, even if lower limits of EF are assumed. The spatial analysis indicates that, although most of attention is being given to vehicle emissions, a significant fraction of Brazilians is exposed to pollutants emitted by stationary sources.
3.036 Future tropospheric ozone impacts of unconventional oil and natural gas development in Mexico.

Early Career Scientist

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Abstract:

New efficient drilling techniques have increased the amount of onshore extractable oil and natural gas (ONG). Consequently, unconventional exploitation of these resources has expanded rapidly into new regions across the U.S. and other countries with proven reserves. Having the sixth largest technically recoverable shale gas resources, Mexico has the potential to increase its current natural gas production by a factor of nine considering the estimated proven natural gas reserves, or by a factor of 270 considering the unproven wet shale gas technically recoverable resources. Studies of U.S. basins have shown that the exploitation of these resources is associated with large and poorly constrained emissions to the atmosphere that include non-methane hydrocarbons (NMHC) and nitrogen oxides (NO\textsubscript{X}). Both species play an important role in tropospheric ozone (O\textsubscript{3}) production through several pathways. By modifying the emissions of NMHC and NO\textsubscript{X}, we designed three different future emission scenarios of ONG production in Mexico to analyze the impact on nation-wide atmospheric composition. The distribution of the NMHC and NO\textsubscript{X} emissions over Mexican territory is based on the geographical distribution of its proven ONG reserves. The total emissions of NMHC are calculated by considering known emission rates per well (kg well\textsuperscript{-1}) using three different well densities (well km\textsuperscript{-2}) associated to each ONG production scenario. Underlying NMHC and NO\textsubscript{X} emissions in the baseline scenario were updated and considered as of the year 2010. For the future scenarios, these emissions followed the same trend from 1999-2010.
3.039 Wintertime NOx Chemistry in Power Plant Plumes.

Early Career Scientist

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Abstract:

Nitrogen oxides (NO\textsubscript{X} = NO + NO\textsubscript{2}) play a key role in atmospheric chemistry and coal-fired power plants are a major source of NO\textsubscript{X} to the atmosphere, making up approximately 30% of emissions in the US (epa.gov). NO\textsubscript{X} emissions can vary seasonally, as well as plant-to-plant, with important impacts on the evolution of the plume chemistry. In particular, due to inefficient plume dispersion, nighttime NO\textsubscript{X} emissions from power plants are held in concentrated plumes, where various reaction and mixing rates can have a strong influence on plume chemistry. During the day, NO\textsubscript{X} catalyzes ozone (O\textsubscript{3}) production, while at night it can react to form nitric acid (HNO\textsubscript{3}) and nitryl chloride (ClNO\textsubscript{2}) and remove O\textsubscript{3} from the atmosphere. These processes are well studied in the summer, but winter measurements are more limited.

We will show results from the aircraft-based WINTER campaign over the northeastern United States, where several nighttime intercepts of power plant plumes were made. The intercepts show variable rates of N\textsubscript{2}O\textsubscript{5} conversion to HNO\textsubscript{3} and ClNO\textsubscript{2}, which, in turn, alters the rate of removal of NO\textsubscript{X} from the atmosphere. Additionally, if the N\textsubscript{2}O\textsubscript{5} conversion is high, then the partitioning between ClNO\textsubscript{2} and HNO\textsubscript{3} formation plays a
significant role in NO\textsubscript{X} removal. Finally, the rate of plume mixing and the background O\textsubscript{3} level also contribute to the rate of NO\textsubscript{X} removal. Through modeling of plume chemistry and dispersion, we will show the relative importance of each of these influences on wintertime power plant plume chemistry and transport and how they influence removal of NO\textsubscript{X} from the atmosphere.
Impacts of flaring emissions in Africa.

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Abstract:
Recent simulations have shown a gap between observed and modeled aerosol optical depths above the Guinean Gulf in West Africa, assuming the possible role of flaring emissions which are really important in this area and not accounted for in this work. It can be expected that these emissions also impact regional air quality and health. In this presentation, recent developments on flaring emission inventories for aerosols and gases obtained from DMSP satellite for the years 1990-2012 and validated over Nigeria will be presented and discussed in terms of temporal and spatial variability. By introducing such emission inventories in RegCM regional climatic model in addition to other anthropogenic sources (traffic, domestic fires, industries, charcoal making, biomass burning..) we will study the relative contributions of the different emissions sources on air quality, climate and health.
Impacts of national Power Development Plan on future air and climate pollutant emissions in Thailand.

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Abstract:

Thailand’s fast growing economic development contributes to a rapid increase of the national energy consumption, leading to about 250 million tons emission of carbon dioxide (CO₂), corresponding to approximately 70% of the overall national greenhouse gas emissions in 2014. At the end of 2015, Thailand released the Thailand’s Integrated
Energy Blueprint (TIEB) 2015-2036 to support the Intended Nationally Determined Contribution (INDC) submitted to the UNFCCC, which stated the national commitment of CO₂ emission reduction of 20-25% during 2020-2030, comparatively to the Business As Usual (BAU). Along with CO₂, energy consumption also emits other climate and air pollutants, such as particulate matter (PM), black carbon (BC), ozone (O₃) precursors, etc. Among the five plans in TIEB, Power Development Plan (PDP) is of prime interest due to the high future demand and planned fuel mix change, consisting in increasing the proportion of clean coal technology to reduce fuel dependence on natural gas. Also, the imported power from neighboring countries is forecasted to be higher, as well as the share of improved renewable energy sources in fuel mix. This would contribute to a significant change in climate and air pollutant emissions.

In order to better understand the future national emission change, we have developed a framework for emission inventory and projection assessment, based on the Greenhouse Gas - Air Pollution Interactions and Synergies (GAINS) model. In this study, emission scenarios generated from TIEB-PDP 2015-2036 will be first described and analyzed. Emission trends of key pollutants of interest including PM, BC, and ozone precursors, especially nitrogen oxides (NOₓ), will be discussed. Finally, impacts of the future emissions under different scenarios using air quality modeling system will be presented and discussed in terms of potential contribution to haze and ozone pollution.
3.047 “Smart” control of emissions from power generation in China and policy implications.

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Abstract:

The power sector is the largest source of CO₂ and many air pollutants. China’s policies of controlling emissions from this sector have been formulated around the goal of reducing total emissions of SO₂ and NOx at the national level by a certain target every five years. Implementation plans for such policies are often vague or lacking, and their effectiveness has been questioned as China faces frequent occurrences of extreme pollution events in recent years. In this presentation, we present a modeling analysis illustrating that large gains in environmental benefits can be achieved by ‘smart’ design of when and where emission reductions of SO₂ and NOₓ should be prioritized, based on atmospheric chemistry and transport fundamentals. Different impact metrics will be considered with a focus on secondary PM₂.₅ and ozone. The model-derived priority regions and periods will be compared with actual emission changes from bottom-up inventory and satellite data, and policy implications will be provided as well.
Recent studies have proposed significant increases in CH\textsubscript{4} emissions from North America over the past decade, and implicated rapid growth in U.S. gas and oil production. The evidence for the increase in North American oil and gas emissions is based on (1) observed increases in co-emitted species such as ethane and propane (2) a trend derived from different atmospheric inversions (3) spatial differences across North America derived from space-based retrievals of column CH\textsubscript{4} abundance. We examine these claims using an ensemble of time-dependent inversions collected as part of the Global Carbon Project, and we also consider what long-term observations from the NOAA aircraft observation network tell us about US emissions. We find that none of the time-dependent inversions estimate large trends in US emissions, and this is true for inversions using only surface observations and for those that use retrieved column CH\textsubscript{4}. Furthermore, we find that short term (< 5 year) trends of up to 1.5 ppb/yr can occur in spatial gradients between the Pacific “background” CH\textsubscript{4} values and continental locations due to transport effects. Furthermore, we show that zonal spatial differences for long-lived atmospheric species are not likely to be sensitive to even large trends due to relatively fast synoptic zonal transport. Finally, we consider the extent to which trends in co-emitted hydrocarbons can be used to estimate emissions of CH\textsubscript{4}. 
Abstract:

$\text{NO}_X$ emissions have significant impacts on both climate and human health due to their contribution to [secondary] $\text{PM}_{2.5}$ and ozone formation. Emissions from many light- and heavy-duty diesel vehicles are currently underestimated by up to an order of magnitude and a factor of 3, respectively. Such underestimates hinder the development and monitoring of efficient air quality policy. In this work, we consider new inventories of real-world $\text{NO}_X$ emissions developed by the International Council on Clean Transportation (ICCT) for 11 vehicle markets (which account for more than 90% of diesel vehicle sales in G-20 countries). In addition, emissions scenarios are developed to model the adoption of stringent diesel $\text{NO}_X$ emissions standards (e.g. US Tier 3 or Euro 6/VI) in these 11 markets by 2040. The GEOS-Chem chemical transport model is used to estimate $\text{PM}_{2.5}$ and ozone concentrations for current baseline emissions estimates and for the alternative scenarios under improved emissions standards. We also evaluate the impact of these concentration changes on global mean surface temperature response and the number of avoided annual $\text{PM}_{2.5}$ and ozone-related premature deaths. Adjoint model calculations are then performed for each of the 11 regions, from which we assess the contribution of grid-scale changes in emissions to these multiple impacts. This allows us to evaluate the extent to which adopting more stringent diesel $\text{NO}_X$ emissions standards in each of the 11 primary modeled vehicle markets influences ozone and $\text{PM}_{2.5}$ exposure burdens both within and outside of these regions. These findings highlight the potential health benefits while also addressing the potential change in global surface temperature due to the tightening of diesel $\text{NO}_X$ emission standards around the world.
Results from a global survey of natural gas flaring from the VIIRS instrument: 2012-2014.

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Abstract:
A global survey of natural gas flaring in 2012-2014 is presented using data collected by the National Aeronautics and Space Administration/National Oceanic and Atmospheric Administration NASA/NOAA Visible Infrared Imaging Radiometer Suite (VIIRS). The accuracy of the flared gas volume estimates is rated at +/- 20%. VIIRS is particularly well suited for detecting and measuring the radiant emissions from gas flares through the collection of shortwave and near-infrared data at night, recording the peak radiant emissions from flares. A total of 17314 individual flare sites were identified with that number steadily growing from 11851 to 13610 per year. The total flared gas volume is estimated at 140 +/- 30 billion cubic meters (BCM) per year, corresponding to 3.5% of global production. While the USA has the largest number of flares, Russia leads in terms of flared gas volume. The largest individual gas flares are observed in Venezuela. Ninety percent of the flared gas volume was found in upstream production areas, 8% at refineries and 2% at liquefied natural gas (LNG) terminals. VIIRS data can provide site- or field-specific tracking of natural gas flaring for use in evaluating efforts to reduce and eliminate routine flaring.
3.053 Short-term variability in natural gas methane emissions from the Marcellus Shale.

Early Career Scientist

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Abstract:
As the objective to decrease methane emissions from the natural gas sector has become an urgent concern, accurate methods to identify ‘super-emitters’ (infrastructure with anomalously high methane emissions that would be economically favorable to mitigate) are critically needed. The Marcellus shale, the most productive natural gas shale field in the United States, has received less intense focus than other basins due to its complex topography and is investigated to provide insights into the characteristics of the distribution of methane emissions.
In July and November of 2015 and June of 2016 over 300 unique well pads were sampled using the Princeton Atmospheric Chemistry Experiment (PACE). This mobile lab includes commercial meteorological, position and methane sensors as well as custom ammonia and ethane sensors. The majority of sites were sampled 1-3 times with selected test sites sampled ~10 times. Several sites that had been successfully sampled previously were resampled several times (5-20) over the course of a day to identify any changes of emission strength.
An inter-comparison of different methodologies including the inverse Gaussian plume method and Large Eddy Simulation modeling was used to provide robust error estimates
related to the methodology. This allows more confidence to be placed on both the
distribution retrieved and trends observed in the data. Specifically, well-level traits that
may be related to the overall emission strength or intermittency of the source strength
were examined. Well production values appeared to be a poor predictor of emissions
strength. Overall ~10% of the sites sampled accounted for ~60% of emissions. Variation
in source strength from single sites were observed within the time scales of a single day
and multiple days that significantly exceeded the uncertainty estimates deduced from
multiple methods. Intermittent plumes may have a direct impact on what is labelled a
super-emitter and implementing appropriate mitigation strategies.
Quantification and Intercomparison of Methane Emissions from Production Sites in Dry Vs Wet Oil and Natural Gas Fields in the Western and Midcontinental United States.

Early Career Scientist

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Abstract:
Over the past decade, natural gas production has skyrocketed in the United States, largely as a result of unconventional methods such as hydraulic fracturing. Studies have shown that national emissions of methane have also increased significantly over the last decade with important climate implications given that methane has a global warming potential of approximately 86 times greater than CO2 over a 20-year period. In this study, methane fluxes were measured at a large number of production sites in four major U.S. oil and gas basins using the Environmental Protection Agency’s (EPA) Other Test Method (OTM) 33a. With this method, measured methane concentrations are combined with Gaussian plume dispersion parameters and concurrent meteorological conditions to obtain a flux estimate. The basins measured in this study range from producing extremely dry to extremely wet natural gas. Dry natural gas contains very little to no condensate, so natural gas extracted from drier basins requires less processing equipment and it has been postulated that fugitive emissions from dry fields will be lower. Measurements were taken in Wyoming’s Upper Green River Basin, Colorado’s Denver-Julesburg Basin, Utah’s Uintah Basin, and a Midcontinent Basin. Mass-weighted averages (MWA) of methane emissions as a percent of production are computed for each basin. These results give a rough estimate of basin-wide emissions from well pads and the computed MWAs for each of the four basins range from 0.14 – 3.11 %, revealing the large variability basin-to-basin. The relationship of leakage rate to how wet or dry a basin is will be discussed along with other potential explanations for the variation in leakage rate.
3.055 Experimental Campaign: Measuring black carbon concentrations in exhaust plumes from heavy duty trucks driving through the Andes Cordillera.

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Abstract:

Emissions from particle matter generated from vehicle activities in high altitude zones (around 3,000 mts.) could increase black carbon deposition over snow and glaciers. A preliminary campaign measuring black carbon concentrations at Los Libertadores highway in Los Andes, Chile, and associated to international freight transportation has been performed during four days in October 2015. A vehicle outfitted with a mini-aethalometer Model AE51 was used, together with a GPS device, to follow heavy duty trucks simulating their driving patterns. This methodology is being tested for a larger campaign to be carried out in the same area, scheduled for July 2016. Differences in black carbon measurements were found from trucks driving to and from the international border, suggesting that diesel quality from Argentina, Bolivia, Paraguay, Brazil and Chile might produce different emission levels. Also, loaded trucks driving uphill show maximum BC levels registered during the campaign. A total of 40 trucks were followed in total, both driving cycles and black carbon concentrations were measured simultaneously. Preliminary results show peaks of 400,000 ng/m³ on trucks operating uphill, background average in zones with no population show values between 2,000 to 3,000 ng/m³ and urban highway values were in average 20,000 to 30,000 ng/m³. Substantial differences on truck exhaust “plume” in relation with other vehicles were observed. Finally, GPS tracking allows correlating emissions results and driving behavior through the Vehicle Specific Power (VSP) methodology. This correlation will be used to model emissions from trucks only knowing their driving pattern, allowing estimating emissions accurately for a longer period of time.

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Abstract:
Non-road diesels account for about a quarter of the nitrogen oxides and primary aerosol emissions from mobile sources in the United States. While a lot is understood about primary emissions, very little work has been done to characterize the atmospheric formation and evolution of secondary organic aerosol (SOA) from tailpipe emissions of non-road diesels. Further, little is understood about how SOA responds to the use of alternative fuels and emissions control systems. To address this gap, in the Summer of 2015, we performed photochemistry experiments on diluted emissions from a 4.5 L John Deere diesel engine using a potential aerosol mass (PAM) reactor. Aerosol size, mass and composition were measured using a scanning mobility particle sizer, photoacoustic extintiometer, and high-resolution aerosol mass spectrometer (HR-AMS). After 1.5 days of simulated atmospheric aging, production of SOA dominated primary aerosol emissions by an order of magnitude. Efficient combustion at higher engine loads and removal of SOA precursors and particle emissions by emissions control systems (oxidation catalyst and particle filter respectively), significantly reduced emission factors for POA (factor of 10-50) and production factors for SOA (factor of 10). The only exception was that the SOA production was nearly identical for idle loads regardless of the use of emissions control systems. We hypothesize (and even validate using carbon monoxide measurements) that at idle loads the oxidation catalyst temperatures were too low to oxidize/remove any of the SOA precursors. Elemental analysis of the HR-AMS data shows a more oxygenated OA at idle than higher loads. Statistically, the use of soy-based biodiesel (B100) produced slightly lower POA emissions but nearly identical SOA levels as diesel. Work is under way to perform a positive matrix factorization using the HR-AMS data and understand compositional differences in OA produced across engine loads, fuels and use of emissions control systems.
Abstract:

The Upper Green River Basin of Wyoming (UGRB) contains two of the top ten gas fields in the U.S., producing approximately 807,386 mcf of natural gas in 2013. This gas is somewhat “wet”, meaning that along with methane, heavier hydrocarbons and water are produced in the extraction process.

A common concern of natural gas production and transmission is the emission of methane (a potent greenhouse gas) and consequently, measurements of fugitive methane from these processes are increasing. Natural gas production also emits many non-methane hydrocarbons (NMHCs), including air toxics such as benzene, toluene, and xylenes, for which little data on emission rates exists. Besides deleterious health effects, these NMHCs are implicated in wintertime ozone production which is difficult to model without accurate emission inventories.

NMHC fluxes are typically estimated via canister grab samples during a methane flux measurement. The methane:NMHC ratio of a canister is determined later in the laboratory, and applied to a known methane flux measured in the field. Utilizing the EPA Other Test Method 33a, a Picarro methane analyzer, and an Ionicon Proton Transfer Reaction-Time of Flight-Mass Spectrometer, the University of Wyoming Mobile laboratory directly measures both methane and NMHC emission fluxes from 38 well pads in the UGRB.

Preliminary results suggest varying NMHC compositions between well pads, disparities between large methane emitters and large NMHC emitters, and multiple NHMC sources on well pads. NMHC fluxes ranging from ~ 0 - 150mg/s have been observed. These results and implications for traditional canister sampling of these sites will be discussed.
3.058 Contrasting winter- and summertime ozone and organic aerosol contributions from the oil/gas sector emissions in the western US.

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Abstract:

Rapid development of oil and natural gas production across the United States in recent years has been associated with significant amounts of methane and other volatile organic compounds (VOCs) released to the atmosphere. The highest ground level ozone (O$_3$) concentrations during 2013 year within the entire country were detected in winter over a remote area, the Uinta Basin (UB) in Utah, which is densely covered by oil and natural
gas wells. During these pollution episodes significant levels of organic aerosol (OA) were also observed. In this study, we contrast wintertime UB O₃ and OA formation with summertime conditions over several shale basins. We use a coupled meteorology-chemistry model, WRF-Chem, to simulate air quality impact of the oil/gas sector emissions in both winter- and summertime conditions. In situ observations from the January-February 2013 UBWOS and June-July 2013 SENEX field studies are used for the WRF-Chem model evaluation as well as for emissions specification for the top-down approach. This method derives VOC and nitrogen oxides (NOₓ) emissions from the oil/gas sector using methane (CH₄) emissions calculated from mass-balance along with ratios of VOC and NOₓ to CH₄ determined from linear regressions. We compare results using this top-down approach with model simulations using the U.S. EPA NEI-2011 (version 2 for the oil/gas sector) national inventory, focusing on O₃ and OA formation. The WRF-Chem model can simulate high O₃ and OA concentrations in the UB during winter of 2013 using the top-down emission estimates. The model shows that oil/gas emissions contribute as much as ~80 ppb to daytime O₃ mixing ratios in the UB during wintertime. However, the WRF-Chem model results show only a small impact on overall summertime O₃ levels from the oil/gas activity emissions. We also discuss the contribution of the oil/gas emissions to secondary OA levels.
3.059 The Influence of BTEX Landfill Gas Emissions: A Case Study on Residents’ in Roodepoort, Gauteng.

Early Career Scientist

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Abstract:

Landfills are known to produce pollutants that have a negative impact on humans and the environment. In developing countries like South Africa, land-filling is the most common form of solid waste management. People living in close proximity to landfills are particularly at risk to harmful pollutants such as the volatile organic compounds (VOC’s). Although VOC’s only constitute about 1% of landfill gas emissions (LFGs), they are of concern as the risk associated with these pollutants is relatively high. Of particular concern, within the VOC’s, is the BTEX group (viz. benzene, toluene, ethyl-benzene and xylenes). These pollutants are wind dispersed to the surrounding residential areas, exposing residents to harmful emissions. Using Radiello Passive Samplers (analysed using a GC with built-in FID) within the residential areas surrounding the landfill, BTEX LFG emissions were quantified and spatially mapped. A 9-day sampling campaign from 27 July to 5 August 2015 was employed. The BTEX levels obtained ranged from 8.83 to 39.62 µg.m⁻³. The effect of the slow south and south westerly wind speeds of 2.44 m.s⁻¹ obtained from the Davis Weather Station positioned in the residential area, the valley terrain, and the location of the landfill resulted in a BTEX ‘hotspot’ location within the Panorama area, north and east of the landfill site. Combined with the sampling strategy, a perception study revealed that respondents’ in this ‘hotspot’ area had concerns regarding the landfill site and its associated pollution. It can be concluded that respondents living closer to the landfill, in the Panorama area, are at higher risk to BTEX emissions, especially during the winter season. This situation thus demands further analysis, specifically during summer, and particularly attention from decision makers. This study should furthermore be considered as a pilot study as many other, more in depth studies may come from this work.
Eight Years of Airborne Methane Measurements in Indianapolis.

Early Career Scientist

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Abstract:
Efforts to better understand anthropogenic and biogenic emissions of methane (CH\textsubscript{4}), a greenhouse gas with a 100-yr global warming potential 25-fold greater than that of carbon dioxide, have become increasingly important as atmospheric CH\textsubscript{4} concentrations have grown to over 2.5 times pre-industrial levels. Landfills and natural gas distribution networks within cities have been shown to be significant CH\textsubscript{4} sources. The Indianapolis Flux Experiment (INFLUX), a multi-institution and multi-method campaign, has been evaluating top-down and bottom-up approaches for quantifying urban greenhouse gas emissions since 2008. The city of Indianapolis is an ideal testbed for studying urban emissions, given its isolation from other major cities and relatively simple topography and meteorology.

Airborne measurements of CH\textsubscript{4} in Indianapolis using Purdue University’s Airborne Laboratory for Atmospheric Research (ALAR), a light twin-engine aircraft instrumented for atmospheric observations, have been ongoing since 2008. To estimate city-wide emission rates of CH\textsubscript{4} from Indianapolis, our group uses a mass balance approach to quantify the city’s contribution of CH\textsubscript{4} relative to background levels. In addition to eight years of city-wide CH\textsubscript{4} emission rate estimates, we will discuss intra- and inter-annual trends in the magnitude of CH\textsubscript{4} emissions from Indianapolis.

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Abstract:

The dual issues of energy and air pollution are increasingly a problem associated with developing countries. While there are a plethora of epidemiological studies that quantify the relationship between mortality and morbidity due to air pollution in North America and Europe (Atkinson et al., 2011; Pope et al., 1995; van der Kamp et al., 2015), there is a paucity of similar data for developing countries. South Africa is a middle-income country that relies on coal for 93% of electricity supply (IRP, 2011). The Vaal Triangle-Highveld region, which includes the Johannesburg-Pretoria megacity, is the largest NO\textsubscript{2} hotspot in the southern hemisphere due to the presence of 11 coal-fired power stations that are not fitted with technologies to reduce NO\textsubscript{2} emissions (or SO\textsubscript{2} emissions), and this small area accounts for 91% of South Africa’s NO\textsubscript{x} emissions (Wells et al., 1996; Lourens et al., 2012). However, the magnitude of the external costs of coal on human health and economic growth in South Africa are as yet unknown. This work aims to provide the first estimates of the health and economic burden of urban air pollution in South Africa, and to quantify the benefits that can be gained from improved air quality. The environmental Benefits Mapping and Analysis Program (BenMAP) model, which systematizes the health impact calculation process, was applied to South Africa using municipality-level census data, air pollutant monitoring data, baseline incidence rates for overall-mortality, and local concentration-response functions (Wichmann et al., 2012). The model was then used to calculate the number of mortalities associated with 1) the current concentrations of NO\textsubscript{2}, SO\textsubscript{2}, and PM\textsubscript{10}, and 2) a reduction of current concentrations to WHO-recommended levels. This study provides the first estimates of the health impacts of urban air pollution at the spatial resolution necessary for establishing cost-effective air quality standards in South Africa.
Emperical characterization of residential solid fuel burning in South Africa's low-income urban areas.

Early Career Scientist

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Abstract:

Solid fuel burning in low-income urban areas is South Africa's biggest air quality problem. Between 40% and 50% of its 53 million people live in areas where solid fuel burning is prevalent. Particulate matter (PM) concentrations are typically between 50% and 100% higher than adjacent urban areas. These emissions are not well represented in current global emission databases and ambient concentrations are poorly sampled using remote sensing due to strong spatiotemporal variability. A struggling economy and large coal-fired power stations that provide more than 90% of electricity create a challenging environment to find solutions to high PM levels in urban areas. Power stations have no SOx, NOx, and aging PM control technologies, nevertheless, regulators are contemplating emission offsetting as a strategy to fund efforts to improve air quality in urban areas. This paper reports on field campaigns that explored different interventions to lower PM emissions in two low-income urban areas where coal burning is reported in 60% to 80% of households. Interventions included thermal insulation, low-emission stoves, electricity subsidies and LPG. The suite of instruments included standard meteorology (Vaisala and RM Young), criteria air pollutants (TEI, Teledyne, Horiba and PICARRO), PM samplers (MetOne E-Bams, MetOne E-samplers, an AE31 7-wavelength Aethalometer MAAP-CBC and a GRIMM), volatile organic compounds (Syntech GC), indoor and household air quality (TSI DustTraks, TSI SidePaks, Meshguard SO2 and CO), as well as indoor and stove temperatures (Thermocron iButtons). Source apportionment was performed in different seasons and sites using coarse and fine nucleopore filters in Stacked Filter Unit samplers and analyzed with energy dispersive X-Ray Fluorescence (XRF) and Ion Chromatography (IC). Drivers of solid fuel burning are identified in order to inform ongoing efforts to design and optimize strategies to reduce emissions.
Study of the air quality in region impacted by emissions from thermoelectric power plants burning sugarcane bagasse in Brazil.

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Abstract:

Brazil is the largest global producer of sugar and ethanol from sugarcane. Ethanol and sugar production generates an excessive amount of bagasse, which is almost all used to generation electricity in thermoelectric power plants, which is approximately of 9.7 GW. The State of São Paulo, region focus of this study, accounts over 60% of the country’s production, occupying approximately half of its area with the cultivation of sugarcane and more than 197 thermoelectric plants burning sugarcane bagasse. Thus, the aim of this study is to evaluate the impact of the pollutants emitted by power plants, which used sugarcane bagasse as fuel, in the region of São Paulo state, which concentrates a large number of power plants, using the model Weather Research and Forecasting with Chemistry. The center of grid is -21.15 and -48.98 with 200 x 150 grid cells and 3 km of horizontal resolution. Preliminary results of the study indicate a significant contribution of this source in air quality in the region, overcoming the contribution of vehicle emissions, mainly for particles and nitrogen oxides pollutants. Moreover, it can be observed the influence of the particles plume in completely South-Southeastern of Brazil. Therefore, the emission from this sector could not be neglected in the regional air quality modeling as well for evaluation of air quality of this area, although are not available inventory for industrial sources in Brazil.
3.067 Effect of shipping emissions on atmospheric composition over the Barents Sea.

Early Career Scientist

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Abstract:

The Arctic is undergoing unprecedented changes as a result of rapid warming and socio-economic drivers. Even though the region is a receptor of anthropogenic pollution from the highly populated mid-latitudes, there are also local sources of pollution, such as shipping, that are already contributing to perturbing atmospheric composition. The Barents Sea, located off the northern coasts of Norway and Russia, has year-round shipping traffic and is likely to grow in a warming Arctic because of the economic benefits related to the opening up of the North-east passage placing it in a strategic position for the transport of goods from Europe to Asia. An increase in the marine traffic has already been observed over the past years in this region, resulting in increased emissions of pollutants.

In this work, carried out as part of EU-FP7 project ICE-ARC (Ice, Climate, Economics - Arctic Research on Change), we study the contribution of the shipping emissions in the Barents Sea on atmospheric composition in the region for the high traffic summer period (July/August) using the regional chemistry-aerosol transport model WRF-Chem run at high resolution over the region. We examine impacts of shipping pollution on production of aerosols, in particular Secondary Organic Aerosol (SOA) and also on deposition (NO$_3^-$, SO$_4^{2-}$) of potentially important nutrients. The model is run using an analytical chemical mechanism for gas phase and aerosols (SAPRC99 coupled with VBS and MOSAIC) for present-day (2012) and future (2050) conditions with ECLIPSE anthropogenic emissions and Winter et al. (2014) shipping emissions. We take into account different future growth scenarios, such as CLE (current legislation) and HGS (high growth scenario), to investigate possible future changes in surface concentrations, total column burden and deposition fluxes. Potential chemistry-climate feedbacks are also examined such as those related to aerosol-cloud interactions or changes in photolysis rates due to shipping.
3.068 Sensitivity of Surface Ozone to Recent Increases of Volatile Organic Compound Emissions from the Growth of North American Oil and Natural Gas Industries.

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Abstract:

After having peaked during 1970-1980 volatile organic compounds (VOC) had been steadily declining in many urban areas and at remote sites in the Northern Hemisphere (NH) mostly due to improved emission controls. In association with reductions of emissions of nitrogen oxides in most of the NH developed nations these atmospheric concentration changes have led to declining, respectively flattening, trends in tropospheric ozone in many densely populated urban and NH remote regions. A reversal of the previously observed declining NH atmospheric trends of light non-methane hydrocarbons occurred around 2009, and NH atmospheric NMHC have since been increasing at rates that are ~5 times faster than the previously observed declines. This increase in atmospheric NMHC has been primarily associated with increased emissions from the rapidly expanding oil and natural gas (O&NG) industry in North America. Emission trends were inferred from observed atmospheric concentrations and applied to the EMAC model to investigate the sensitivity of regional and large-scale changes in tropospheric ozone to the increased NMHC emissions. We find an average summer increase in surface ozone of up to 2.5 ppb near O&NG source regions from 2009 - 2014, and increases of >0.5 ppb extending far downwind over the North Atlantic, North Africa, and Middle East region. Furthermore, this added ozone production adds to occurrences of exceedance of the 70 ppbv National Air Quality Standard (NAAQS) in numerous U.S. non-attainment regions.
3.069 Global methane budget and natural gas leakage based on long-term δ13C-CH4 measurements and updated isotopic source signatures.

Early Career Scientist

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Abstract:

Quantifying and mitigating CH4 emissions associated with extraction and use of fossil fuels (natural gas, oil, and coal) has been the focus of many measurement U.S. field campaigns, modeling studies, and policy and industry activities in recent years. While this enhances the information available to better understand the impact of fossil fuels on U.S. CH4 emissions, global modeling studies frequently suffer from limited data to attribute total CH4 emissions to individual sources. We generated isotopic source signature distributions based on the largest literature survey to date to better constrain global fossil fuel CH4 emissions. These are combined with atmospheric measurements from NOAA’s Global Greenhouse Gas Reference Network and the literature spanning the past three decades including globally averaged CH4 and stable isotopes (13CCH4). A Monte Carlo box-model and a global transport model were used to estimate distributions and confidence intervals of individual emissions sources. Attributing the majority of increased CH4 levels over the past three decades to microbial sources is consistent with 13CCH4 records. The sum of CH4 emissions from fossil fuel extraction and use and geological seepage is significantly larger than previous estimates. Finally, recently published estimates of global CH4 emissions from the oil and coal industries are subtracted from our global fossil fuel CH4 results to quantify global CH4 leakage from the natural gas industry during extraction, processing, transport, and distribution. Natural gas CH4 leakage as a fraction of total production has decreased steadily over the same period indicating industry efficiency improvements.
High wintertime ozone in the Uinta Basin: Insights from aircraft, tethersonde and surface measurements.

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Abstract:

Extreme high ozone mixing ratios, far exceeding U.S. National Air Quality Standards, were observed in the Uinta Basin in January-February 2013. Extensive surface and tethered balloon profile measurements of ozone and meteorological variables as well as aircraft measurements of O₃, CH₄, CO₂, NO₂ and a suite of non-methane hydrocarbons (NMHCs) link emissions from oil and natural gas extraction and production activity with strong ozone formation. Under conditions highly favorable for wintertime ozone production including snow-covered ground, prolonged temperature inversions, and air confining topography, hourly average ozone mixing ratios built up from regional background levels of 40-50 ppbv to >160 ppbv during several multiday episodes. High ozone mixing ratios were found across the entire Basin and extended from the surface to the top of the inversion layer at ~200 m above ground level. This layer was at a nearly uniform height across the Basin even though there are significant terrain variations. Very high mixing ratios of CH₄ were also seen throughout the Basin within the inversion layer. High levels of NMHCs that were highly correlated with CH₄ showed that abundant O₃ precursors were available throughout the Basin leading to Basin-wide production. Ozone levels were well correlated with CH₄ implying that the associated NMHCs were key precursors in ozone formation. The highest CH₄ and NMHC levels were found near the main natural gas field in the central to eastern portion of the Basin. Surface wind data showed a consistent diurnal pattern driven by heating on the rim of the Basin that moved air from the vicinity of the gas field to the edges of the Basin during the day with air draining back into the Basin at night. This transport pattern was likely responsible for dispersing ozone and its precursors throughout the Basin.
3.071 Assessing the impact of the Martin Drake coal-fired power plant on ambient mercury concentrations in Colorado Springs, CO, USA.

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Abstract:

Mercury (Hg) is a bioaccumulative neurotoxin emitted from both natural and anthropogenic sources. Due to the complex chemistry that Hg undergoes in the atmosphere, the spatial impacts of Hg emissions range from local to global in scale. As Hg is primarily introduced to terrestrial and aquatic ecosystems through atmospheric deposition, characterizing the emissions from individual sources is paramount to effectively mitigating ecosystem Hg contamination. Presently in the U.S., coal combustion for electricity generation is the largest source of Hg to the atmosphere. Colorado Springs is home to the Martin Drake coal-fired power plant (CFPP), which provides a significant fraction of the electricity for the local community but has been the center of much scrutiny due to increasing concerns over the age of the plant, its impact on local air quality, and emissions of greenhouse gases that contribute to global warming. While monitoring and modeling efforts have considered the plant’s emissions of criteria air pollutants such as sulfur dioxide (SO$_2$), there has to our knowledge been no active monitoring of hazardous air pollutants, such as Hg. To quantify Hg concentrations in the ambient atmosphere and assess the impact that this CFPP has on the local environment, during summer 2016 we continuously measured total gaseous Hg (TGM) and carbon dioxide (CO$_2$) at a site approximately one mile north-northwest of the plant where SO$_2$, carbon monoxide (CO), and meteorological parameters are also continuously measured. We use this data to elucidate the dominant chemical and meteorological signatures associated with enhancements in ambient Hg. We augment these measurements with air mass transport modeling using the HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model to identify the major source locations associated with observed Hg enhancements, and to determine whether the Martin Drake CFPP emissions significantly elevate ambient Hg concentrations in Colorado Springs.
3.072 Simulating the contribution of emissions from oil and gas development to regional nitrogen deposition at National Parks within the Intermountain West.

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Abstract:

Over the last ten years, oil and natural gas extraction has rapidly increased in the Intermountain West states of Wyoming, Colorado, Utah and New Mexico. These operations often occur near sensitive wilderness areas and national parks. Ecosystems within these areas are often near or above a "nutrient nitrogen critical load", meaning that additional nitrogen deposition may foster unwanted changes in plant communities. This study uses the 2011 WAQS (Western Air Quality Study) CAMx (Comprehensive Air Quality Model with Extensions) platform to simulate nitrogen deposition at several western National Parks, and to determine the contribution of emissions from oil and gas sources to this deposition estimate. A fine scale (4km) grid was employed to assess within-park deposition gradients. A detailed emission inventory was developed for the WAQS, including a comprehensive survey of the oil and gas development sector. CAMx predicts both wet and dry deposition of a full suite of nitrogen-containing gases and particles, including ammonia (NH3), nitric acid (HNO3), nitrogen oxides (NOx), peroxycetyl nitrates (PAN), and particulate ammonium (NH4+) and nitrate (NO3-). National Park Units that were significantly affected with regard to nitrogen deposition from oil and gas sources include Mesa Verde National Park in southwestern Colorado (0.61 kg N/ha/yr, 19% of total deposition), Dinosaur National Monument in northeastern Utah/northwestern Colorado (0.58 kg N/ha/yr, 20% of total deposition), Hovenweep National Monument in southeastern Utah/southwestern Colorado (0.43 kg N/ha/yr, 14% of total deposition), and Rocky Mountain National Park in northern Colorado (0.26 kg N/ha/yr, 8% of total deposition). The bulk of the impact is in the form of oxidized nitrogen deposition, suggesting that NOx controls for oil and gas development equipment (e.g., drill rigs, compressor engines) would be most effective for reducing harmful downwind impacts at National Parks.
Abstract:

Aerosol observations above the Southern Ocean and Antarctic sea ice are scarce. Measurements of aerosols and atmospheric composition were made in East Antarctic pack ice on-board the Australian icebreaker *Aurora Australis* during the spring of 2012. One particle formation event was observed during the 32 days of observations. This event occurred on the only day to exhibit extended periods of global irradiance in excess of 600 W m⁻². Within the single air-mass influencing the measurements,
number concentrations of particles larger than 3 nm (CN₃) reached almost 7700 cm⁻³ within a few hours of clouds clearing, and grew at rates of 5.6 nm h⁻¹. Formation rates of 3 nm particles were in the range of those measured at other Antarctic locations at 0.2–1.1 ± 0.1 cm⁻³ s⁻¹. Our investigations into the nucleation chemistry found that there were insufficient precursor concentrations for known halogen or organic chemistry to explain the nucleation event. Modelling studies utilising known sulfuric acid nucleation schemes could not simultaneously reproduce both particle formation or growth rates. Surprising correlations with Total Gaseous Mercury (TGM) were found that, together with other data, suggest a mercury driven photochemical nucleation mechanism may be responsible for aerosol nucleation. Given the very low vapour pressures of the mercury species involved, this nucleation chemistry is likely only possible where pre-existing aerosol concentrations are low and both TGM concentrations and solar radiation levels are relatively high (‘1.5 ng m⁻³ and < 600 W m⁻², respectively), such as those observed in the Antarctic sea ice boundary layer in this study or in the global free-troposphere, particularly in the Northern Hemisphere.
4.005 OH-Radical Oxidation of Organic Species at the Air-Water Interface.

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Abstract:

Products and intermediates of the prompt oxidation of a variety of carboxylic acids, including cis-pinonic acid and benzoic acid, initiated by gas-phase hydroxyl radicals, \( \times \text{OH}(g) \), at the air-water interface were detected by mass spectrometry in a novel setup under various experimental conditions. Exposure of sub-millimolar reactant aqueous microjets to \( \sim 10 \) ns \( \times \text{OH}(g) \) pulses from the 266 nm laser flash photolysis of \( \text{O}_3(g)/\text{O}_2(g)/\text{H}_2\text{O}(g)/\text{N}_2(g) \) mixtures yielded an array of interfacial species, including peroxyl radicals \( \text{ROO}\times \) and hydroperoxides \( \text{ROOH} \), that were unambiguously and simultaneously identified \textit{in situ} by mass spectrometry. Our results are consistent with a Langmuir–Hinshelwood mechanism rather than an Eley-Rideal mechanism. We propose an interface-specific oxidation mechanism for the heterogeneous reaction of amphiphilic organic species (aq) with \( \cdot \text{OH}(g) \). The connection between the present fundamental findings and atmospheric chemistry is discussed.
4.006 Quantification of gas-wall partitioning in Teflon environmental chambers using rapid bursts of low-volatility oxidized species generated in-situ.

Early Career Scientist

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Abstract:
Partitioning of gas-phase organic compounds to the walls of Teflon environmental chambers is a recently reported phenomenon that can affect the yields of reaction products and secondary organic aerosol (SOA) measured in laboratory experiments. Reported timescales for reaching gas-wall partitioning (GWP) equilibrium ($t_{GWE}$) differ by up to three orders of magnitude, however, leading to predicted effects that vary from substantial to negligible. A new technique is demonstrated here in which semi- and low-volatility oxidized organic compounds (saturation concentration $c^*<100 \mu g m^{-3}$) were photochemically generated in rapid bursts in-situ in an 8 $m^3$ environmental chamber, and their decay due to loss to the walls in the absence of aerosol was measured using a high-resolution chemical ionization mass spectrometer (CIMS) equipped with an “inlet-less” NO$_3^-$ ion source. Measured $t_{GWE}$ were 7-13 min for all compounds, with an average relative standard deviation of 33% for replicate experiments. The fraction of each compound that partitioned to the walls at equilibrium follows absorptive partitioning theory with an equivalent wall mass concentration in the range 0.3-10 mg $m^{-3}$. By comparison, $t_{GWE}$ measured using a CIMS equipped with a standard low-pressure ion-molecule reaction region were biased high by up to a factor of 4, and the equivalent wall mass concentrations were biased low. Thus, improved ion sources that reduce the contact of compounds with walls are needed for low-pressure CIMS. On the basis of these results, a set of parameters is proposed for modeling GWP in chamber experiments. We applied these findings to fit the results of an ISOPOOH oxidation study in the Caltech chamber in which we reported a new SOA formation mechanism. The effects of relative humidity and temperature on gas-wall partitioning, and wall losses on SOA experiments in which competing aerosol seed particles are generated in the chamber were also explored.
4.008 Temperature dependence of bromine activation due to reaction of bromide with ozone in a proxy for aerosols and sea ice.

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Abstract:

The discovery of boundary layer ozone depletion events (ODEs) in the Polar Regions and in the mid-latitudes, two areas of very different temperature regimes, begs the question of temperature dependence of reactions responsible for these observations. These ODEs have been attributed to ozone reacting with halides leading to the formation of reactive halogen species (halogen activation) of which bromide is extensively studied, R1 – R3 (“mp” means multiphase reaction).

O3 + Br→ O2+ OBr⁻   mp (R1)
OBr⁻ + H⁺ ↔ HOBr           (R2)
HOBr + H⁺ → Br₂ + H₂O     (R3)

Despite extensive studies of ozone-bromide interactions in different media, the temperature dependene of and the role of the matrix on bromide activation are not clear; this limits the parameterization of the relevant processes in atmospheric models. In addition, properties of the matrix such as diffusivity (and viscosity), and gas solubility, which are also temperature dependent are not well characterized for binary and complex matrices involving both organics and inorganics, and at low temperatures (such as in aerosols, snow, or sea ice). In bromide-ozone systems, these properties influence the reactive uptake of ozone.

With the application of flow tubes, the aim of this study is to investigate the temperature dependence of bromine activation via ozone interaction in a bromide containing film. Citric acid was used as a hygroscopically characterized matrix in the film, which is of relevance to atmospheric chemistry.

We present reactive ozone uptake measured between 258 and 289 K. The data show high reproducibility. With available knowledge, we have compared the measured uptake with modelled bulk uptake while accounting for temperature dependence of the substrate’s properties. The extrapolated model parameters are discussed.
4.009 Evaluation of Ozone Depleting Substance Replacement Compounds: OH rate coefficients, infrared spectrum, and global warming potentials of (E)-(CF3)2CFCH=CHF (HFO-1438ezy(E)).

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Abstract:
The Montreal protocol and its amendments and adjustments have regulated the phase-out of ozone depleting substances (ODSs) due to their adverse effects on the stratospheric ozone layer. In parallel to the phase-outs, ODS replacements have been proposed that are intended to be less harmful to the environment following their release into the atmosphere during production and use. Hydrofluoroolefins (HFOs) represent the “next generation” of replacement compounds in applications such as fire suppression, heat transfer, blowing, and refrigeration. HFOs are attractive replacement compounds because the presence of the carbon-carbon double bond leads, in general, to a short atmospheric lifetime, which is a key metric in determining their environmental and climate impact. Laboratory evaluations of a replacement compounds atmospheric loss processes is, therefore, a key element in the evaluation of the environmental suitability of a replacement compound.

In this work, rate coefficients for the gas-phase reaction of the OH radical with (E)-(CF3)2CFCH=CHF ((E)-1,3,4,4,4-pentafluoro-3-(trifluoromethyl)-1-butene, HFO-1438ezy(E)) were measured using a pulsed laser photolysis-laser induced fluorescence technique (PLP-LIF) over a range of temperature (214–380 K) and pressure (50–450 Torr, He or N2 bath gas) and with a relative rate method at 296 K at pressures between 100 and 400 Torr (synthetic air). The atmospheric lifetime of HFO-1438ezy(E) was estimated to be ~36 days with respect to OH reactive loss and it is thus considered a very short-lived substance (VSLS). Its actual lifetime will, therefore, depend on the time and location of its emission. The infrared absorption spectrum of HFO-1438ezy(E) was also measured as part of this work. In addition to the experimental measurement results, the radiative efficiency, global warming potentials (GWPs), and photochemical ozone creation potential (POCP) of HFO-1438ezy(E) will be presented.
Development of Portable Chemosensors for Atmospheric Radicals.

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Abstract:

The complex chemical photo-oxidation cycles involved in the degradative removal of anthropogenic and biogenic hydrocarbons from the atmosphere are mediated by radical intermediates. Peroxy radicals (HO$_2$ and RO$_2$) are key reactive intermediates/chain propagators directly involved in the formation of ground-level ozone, photochemical smog and in the production of secondary organic aerosols. Thus they can significantly impact upon air quality and human health.

Speciated measurements of atmospheric radicals pose considerable challenges to analytical chemistry. Owing to their low concentrations, high reactivity, and short lifetimes, free radicals cannot be easily sampled and hence direct offline analysis is extremely difficult. Although a number of highly sensitive sophisticated techniques have been developed, selectivity, full structure determination, portability and cost remain challenging obstacles to atmospheric radical analysis.

We present the synthesis and development of a series of novel chemosensors, organic trapping compounds that are stable and can efficiently and selectively react with a range of important atmospheric radical species. The reaction products conserve the radical structure and are stable enough for off-line analysis using a range of EPR and mass spectrometric techniques. This approach allows accurate determination of the radical structures.

Modification of sample supports has also been investigated, leading to a greater reactive surface area, giving a better sampling efficiency and increasing the temporal resolution of measurement.

The developed chemosensors have been tested and evaluated in laboratory and chamber experiments using a range of atmospherically relevant systems (e.g. ozonolysis of small alkenes and monoterpenes, reactions of OH with alkanes), giving key insights into radical selectivity and reaction mechanisms.
4.012 A protocol for the prediction of photolysis processes in numerical models.

Early Career Scientist

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Abstract:

Chemical mechanisms are essential for our understanding of air quality and climate change, which have a significant impact on society and economy. A benchmark mechanism is needed to study fundamental chemical processes and assess the quality of simplified mechanisms for large-scale modeling. A widely used benchmark mechanism is the Master Chemical Mechanism (MCM, http://mcm.leeds.ac.uk). With ~17,000 reactions, a current research focus is on the automation of the mechanism generation process and the design/update of a protocol for mechanism self-generation.

Photolysis processes contribute to some of the largest uncertainties in the mechanism. This comprises both uncertainties in laboratory measured cross sections and quantum yields for single compounds and in the methodology for estimating photolysis parameters for species without laboratory measurements. We have extended the TUV5.2 photolysis model to include 147 species leading to 240 photochemical pathways.
The extended TUV model is used to update the MCM methodology for determining the photolysis rates for compounds without laboratory measurements. Recent laboratory studies allow us to provide new relationships for some classes of compounds including carbonyls, organic nitrates and hydroperoxides. However, for compounds with multiple absorbing functional groups (chromophores) the laboratory dataset is extremely limited (6 compounds in the current database). These oxidized materials play a central role in the atmospheric degradation of organics and so in the production of secondary organic material and ozone; however, the lack of laboratory data makes the evaluation of their photolysis extremely difficult. We present modeling simulations of the impact of different choices of newly developed parameterizations of the photolysis of these compounds and their effects on the chemistry in different airmasses.
4.013 Synthesizing laboratory and field measurements to trace the bulk oxidation properties and volatility distributions of gas-phase species detected by chemical ionization mass spectrometry.

Early Career Scientist

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Abstract:

We utilize multiple laboratory and field high-resolution time-of-flight chemical ionization mass spectrometry (HR-ToF-CIMS) datasets to assess the volatility distribution and bulk oxidation properties of the identified gas-phase. We find that HR-ToF-CIMS utilizing multiple reagent ions detects species across a large volatility distribution, with species categorized as intermediate-volatility organic compounds (IVOC) dominating the total signal and most influencing the bulk O:C and H:C ratios. We also find that preliminary results in bulk oxidation properties can be extended to differences in aerosol yields of a variety of biogenic volatile organic compounds (BVOCs). Datasets used in this study include small and large chamber experiments of the photo-oxidation of a variety of BVOCs as well as measurement campaigns from sites largely influenced by biogenic sources (southeast US) and a combination of biogenic and anthropogenic source influences (Colorado Front Range). We calculate bulk O:C and H:C ratios for each dataset as a function of oxidant concentrations and ambient conditions. We further determine the volatility distributions of the identified gas-phase species by estimating their vapor pressures based on assumed structure-activity relationships. The calculated vapor pressures were separated into traditional volatility bins, including semi-volatility organic compounds (SVOC), IVOC, and low-volatility organic compounds (LVOC). We determine by signal attribution which volatility bins are most impacting the bulk O:C and H:C values and how the O:C and H:C ratios for each volatility bin change with photo-oxidation and ambient conditions. The bulk oxidation properties of the gas-phase were further extended to aerosol formation and growth in the laboratory and field studies in order to develop a preliminary predictive capability of aerosol yields as a function of the bulk oxidation properties of the precursor gas-phase species.
4.016 Development of a Global Atmospheric Chemistry Model for the Study of PAHs (EMAC-PAH) and Application.

Early Career Scientist

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Abstract:

The ECHAM/MESSy Atmospheric Chemistry (EMAC) model has today become a more favorable tool to simulate global tropospheric and stratospheric chemistry and climate. MESSy provides a framework for the interconnection of the base model (i.e., ECHAM5) and earth-system components, i.e., sub-models describing atmospheric composition, processes and its interaction with oceans, land and human activities. This framework makes the integral model very flexible for use in a wide variety of applications and its development has been continuous. In this work, we expanded the model to include simulations of the transport and fate of polycyclic aromatic hydrocarbons (PAHs) and other semi-volatile contaminants. These substances persist in environmental compartments and pose a health hazard and bio-accumulate along food chains. EMAC-PAH takes into account essential environmental processes including (1) gas-particle partitioning, (2) dry and wet depositions, (3) chemical and biotic degradations, and (4) volatilizations from ocean, land, vegetation, and snow. The gas-particle partitioning is based on a dual approach parameterization, assuming absorption into organic carbon (OC) and adsorption to black carbon (BC) aerosols. We selected four compounds for study, spanning a wide range of volatility, i.e., phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), and benzo[a]pyrene (BaP). Simulations were carried out to study the significance of homogeneous and heterogeneous oxidations. Observation data from multiple land monitoring networks, including the Arctic, and two ship measurements during 2007-2009 were employed to evaluate the model performance.
4.021 Solid versus liquid phase state and glass transition temperature of secondary organic aerosols in the global atmosphere.

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Abstract: 

Secondary organic aerosol (SOA) can adopt amorphous solid, semisolid or liquid phase states depending on chemical composition, relative humidity and temperature. The phase state plays a critical role in SOA formation and evolution as well as activation to cloud droplets and ice crystals. We developed a parameterization to estimate glass transition temperature of SOA as a function of molar mass and O:C ratio of SOA compounds, which can be applied in chemical transport models for the prediction of SOA phase state. Global maps of average molar mass, glass transition temperature and phase state of SOA were simulated using the atmospheric chemistry general circulation model. Global simulations show that the occurrence of amorphous solid or semi-solid phases is a general phenomenon, especially over dry areas. Liquid phase state is expected in the region with high relative humidity such as in the Amazonian and other tropical forests. Vertical simulation results show that the occurrence of solid phase increases at higher altitudes; almost all SOA particles are expected to be solid at 500 hPa.
4.022 Investigation of the below-cloud scavenging of the soluble inorganic aerosols by the rainfall during summer in Beijing.

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Abstract:

In order to determine the relationship between the below-cloud scavenging and the precipitation intensity, high resolution of the aerosol concentration and the evolution of the chemical composition in precipitation through each event were observed in Beijing during 3rd Jun and 4th Jul 2014. It is the first field measurement study which focus on the below-cloud scavenging coefficients of the soluble inorganic aerosols based on the sequential sampling method in Beijing. Averaged concentration of $\text{NO}_3^{-}$, $\text{SO}_4^{2-}$, $\text{NH}_4^{+}$ in the precipitation were 158.3, 205.7, 338.6 μeq/L respectively, while the below cloud scavenging is much more significant than that in cloud process with about 2/3 fractions to 1/3. Eliminated the cleaning up effect of the north wind, the below cloud scavenging is calculated by the below-cloud scavenging ratio multiplied by the precipitation intensity and divided by the cloud-base height. The relationship between the below-cloud scavenging efficiency and the precipitation intensity is determined by the exponent function $K=aP^b$. For $\text{NO}_3^{-}$, $\text{SO}_4^{2-}$ and $\text{NH}_4^{+}$ the relationship is $K=4.79\times10^{-3}p^{0.75}$, $K=1.28\times10^{-3}p^{1.20}$ and $K=8.45\times10^{-4}p^{1.20}$, respectively. Compared to the literature reported over the world, the parameter $a$, which depends on the precipitation type and the aerosol particle size, is at the same order of magnitude. Due to the high aerosol concentration in the air mass and short-time rainstorm events frequently occurred in summer of Beijing the parameter $b$ obtained in this study is a little higher than the others, which indicated the soluble inorganic aerosols were more convenient to be scavenged from the atmosphere. Therefore, this study offered a new opportunity to improve the wet scavenging procedure in the 3-D numerical model and hence the simulation of the wet deposition in Beijing.
Improved analyzer for biogenic volatile organic compounds as total ozone reactivity and its application to kinetics of gas-phase reactions.

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Abstract:

Biogenic volatile organic compounds (BVOCs) have been focused on as precursors of tropospheric ozone (O_3) and secondary organic aerosols. Various species of BVOCs have C=C double bonds and can react with O_3. To capture BVOCs comprehensively, a total ozone reactivity (R_O3, the sum of k_i[VOC_i]) analyzer has been developed [1-3]. R_O3 of sample BVOCs can be determined when decrease of O_3 due to BVOCs+O_3 is precisely monitored. Initial O_3 concentration was set to be 30 ppbv. Cyclohexane was added as an OH scavenger. Detection limit of the prototype analyzer was 1.4x10^{-4} s^{-1} (S/N=3, 60-s average, 60-s reaction). To apply the analyzer to field observations and/or laboratory studies where precise measurements of O_3 and R_O3 are essential, further improvement of the analyzer is necessary. In this study, the analyzer was successfully improved through optimization of (1) O_3 detector, (2) O_3 supply, and (3) reaction chamber. The acquired detection limit of R_O3 was 0.2x10^{-4} s^{-1}. As an application, temperature dependence of rate constant, k(T), for gas-phase reaction of a BVOC with ozone was explored. In this study, linalool, (CH_3)_2C=CHCH_2C(CH_3)(OH)CH=CH_2, was examined. For linalool, k(296K) can be referred as 4.3x10^{-16} cm^3 molecule^{-1} s^{-1} [4]. The detection limit of improved R_O3 analyzer corresponded to 5 ppbv as linalool equivalent. R_O3 of linalool at 310 K was quantified as 5 % smaller than that at 296 K. k(310K) was estimated as 4.1x10^{-16} cm^3 molecule^{-1} s^{-1}. Consequently, it was indicated that the rate constant of linalool+O_3 reaction decreased by 5 % when the temperature changed from 296 K to 310 K.

4.025 Diffusivity measurements of volatile organics in levitated viscous aerosol particles.

Early Career Scientist

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Abstract:

Field measurements indicating that atmospheric secondary aerosol particles can be present in a highly viscous, glassy state [1] have spurred numerous studies addressing kinetic limitations of hygroscopic growth due to slow condensed phase diffusion and measurements of water diffusivities in viscous organics [2, 3]. In contrast, little is known about diffusion limitations of organic molecules in viscous matrices and how these might affect gas-particle phase partitioning of complex mixtures with constituents of different volatility, the questionable assumption often being that viscosity data can be used to infer diffusivities according to Stokes-Einstein.

In this study, we developed a measurement procedure and provide a quantitative estimate for the diffusivity of a volatile organic in a viscous matrix. Single particles generated from an aqueous solution of sucrose and a small quantity of volatile polyethylene glycol (PEG) are investigated in an electrodynamic balance at controlled humidity conditions over a large temperature range, thereby varying the viscosity of the sucrose matrix. Mie resonance spectroscopy is used to determine the particle size and evaporation rate. At high temperatures, no diffusion limitations apply and the particle remains well-mixed throughout the evaporation process. In the kinetically limited regime at low temperatures, the surface concentration of PEG is depleted as the diffusion flux emanating from the center of the particle is too low to maintain a homogeneous concentration throughout the particle. Therefore, we observe a deviation of the evaporation rates from the temperature dependence as predicted by the pure component enthalpy of vaporization under well-mixed conditions. The resulting reduced evaporation rates are used to determine the PEG activity at the surface and to quantify the PEG diffusivity.

4.026 Phase transition in salt solutions at the bulk and interface with core level spectroscopy.

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Abstract:

Sea salt, and in particular chloride, is an important reactant in the atmosphere. Chloride in sea salt aerosol is – once chemically converted to molecular halogens (Cl2, BrCl) and released to the atmosphere – well known as important atmospheric reactant. A crucial factor determining the overall reactivity of this halogen release is the local physical environment of the chloride ion. For example, the reactivity of liquid aerosols decreases significantly upon crystallization. Surprisingly, the phases of NaCl-water systems are still under debate. Using core electron spectroscopy of the oxygen atoms in water, we previously showed that these systems follow the phase rules at the air-ice interface. This finding contrasts some earlier observations, where the presence of liquid below the eutectic point of bulk solutions was postulated.

In the present study, we present new electron yield near-edge X-ray absorption fine structure spectroscopy (NEXAFS) data obtained at near-ambient pressures up to 20 mbar of NaCl frozen solutions. The method is sensitive to small changes in the local environment of the chlorine atom and can either probe the surface or the or bulk of the sample. The study indicates frapant differences in the phases of NaCl - water mixtures at temperatures below the freezing point for the surface of the ice vs. the bulk. This has significant impact on modelling chemical reactions in snow or ice and it’s environmental consequences. Further, this study reveals more general and new insight into the fundamental question of how the local environment of dopants change upon freezing at the air-ice interface. In this presentation I will thus also focus on presenting our new set-up to allow to precisely control temperature and pressure for sub-freezing core electron spectroscopy studies.
4.027 Mass-independent isotopic fractionation and transport in volcanic sulfates.

Early Career Scientist

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Abstract:

Atmospheric sulfates play an important role in the Earth’s radiative balance [IPPC report, 2013]. Volcanoes are among the main natural sources of sulfur in the atmosphere. Once injected in the atmosphere, volcanic sulfur is oxidized and converted to sulfate particles. There are still significant uncertainties pertaining to the atmospheric sulfur oxidation in different regions. Mass-independent isotopic fractionations (MIF) of sulfur oxidized species can be used as quantitative markers of oxidation pathways in the atmosphere and to constrain the sulfur cycle. The majority of the oxygen-MIF anomalies observed in sulfur species originate from the ozone anomaly transferred during oxidation, while the majority of the sulfur-MIF anomalies come from the photochemistry of atmospheric sulfur species.

The isotopic composition of sulfate in volcanic deposits has been measured in different regions of the world. We present here the analysis of the isotopic data using a photochemical box-model (CiTTyCAT) that contains a detailed description of tropospheric chemistry. The box-model can be further coupled to a Lagrangian model (FLEXPART) to simulate the transport of chemically evolving air masses. We have added a detailed sulfur chemistry scheme which includes its heterogeneous chemistry in the aqueous phase. Moreover, we have implemented and linked to it an isotope sulfur oxygen scheme, which allows to model the time evolution of the oxygen-MIF in volcanic sulfates during the oxidation of volcanic sulfur. The MIF anomalies modeled in volcanic sulfate are finally compared to the isotopic measurements made on sulfates collected in different regions.
Chemistry-clouds interactions over West Africa: the role of moist thermals on the atmospheric oxidation capacity.

Early Career Scientist

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Abstract:

In the framework of the EU-funded (FP7) DACCIWA project, this work aims at quantifying the impact of turbulent mixing on the chemical reactivity in the atmospheric boundary-layer (ABL). The Large-Eddy Simulation version of the french model Meso-NH is used which explicitly resolves the turbulent and convective advection terms. The effect of homogeneous emissions representative of biogenic environment and mixing induced by thermals on the redistribution of chemical species is assessed in a convective ABL. Typical dynamic conditions during the monsoon period over coastal West Africa are considered for this study. The chemical reactions are calculated on-line with a detailed chemical scheme describing reactions of ozone and SOA gaseous precursors. The objective is to estimate the impact of mixing by thermals on the spatial distribution and segregation of chemical reactants and to understand the contrasted oxidizing capacity inside versus outside of the thermals. A focus is made on the oxidation reaction of isoprene by the OH radical, which chemical timescale is much shorter than the turbulent timescale. A reduction of almost half of the reaction rate is obtained due to the segregation of reactive compounds on the top of the boundary-layer compared to unperturbed, non turbulent conditions. Additional effects of spatial heterogeneous emissions including anthropogenic emissions and aqueous phase chemistry are discussed.

Early Career Scientist

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Abstract:

By examining the differences in simulated PM_{2.5} and O_3 and associated uncertainties, we quantitatively explore potential trade-offs between complex, high-resolution, short simulations and simpler, coarse-resolution, long simulations for air pollution health impacts analysis. Continuing advancements in computational power, observations, and scientific understanding allow for more complex simulations with higher spatial resolutions. This enhances our ability to assess human health hazards due to atmospheric chemistry and climate change, but also increases awareness of parametric uncertainties within models and structural uncertainties between models. Thus, analysts often face a choice between shorter or fewer runs of a complex model and longer or more runs of a simpler model. This work explores the implications of model choice for health benefits analysis using two state-of-the-art chemistry-climate models with a particular focus on O_3 and PM_{2.5} exposure. We examine differences resulting from modeling decisions regarding: resolution, chemical mechanism, driving meteorological data, and model choice. Preliminary comparisons of CESM CAM-Chem simulations using standard CESM CAM-Chem and GEOS-Chem emissions show that, when normalized by their respective mean values, global O_3 shows twice as much of a difference between models than global...
The opposite is found over the US, where the differences between models for O$_3$ are half as much as differences between models for PM$_{2.5}$. We also find that CESM simulations, when comparing standard CESM CAM-Chem and GEOS-Chem emissions, result in time-series correlations that are higher globally than regionally (e.g. PM$_{2.5}$ correlations of $R^2=0.99$ globally, and $R^2=0.43$ over the US). Preliminary comparisons of simulations using chemical mechanisms with differing complexity find small differences for aerosols ($\sim 10\%$) and larger differences for gaseous species such as O$_3$. Uncertainties related to resolution and driving meteorological data may be of comparable magnitude. Finally, we assess the influence of these choices on health prediction using the BenMAP health impacts assessment software.
4.032 Molecular Halogens in the Arctic: Fluxes, Diurnal Variations, and Implications.

Early Career Scientist

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Abstract:

High levels of reactive bromine and chlorine species have been frequently observed in the Polar boundary layer, which lead to ozone depletion events and mercury oxidation and deposition. Recent studies reported Arctic snowpack photochemical production of Br\(_2\) and Cl\(_2\), yet mechanisms remain poorly understood and largely untested. In this work we use a zero-dimensional multi-phase model to examine the reactive bromine and chlorine chemistry observed during the 2012 BRomine, Ozone, and Mercury EXperiment (BROMEX), focusing on the production and removal of molecular halogens near Barrow, Alaska. The roles of primary snowpack production, gas-phase recycling, and heterogeneous recycling are examined for Br\(_2\) and Cl\(_2\) observations. Nitrogen-containing halogens, such as ClONO\(_2\), BrONO\(_2\) and BrNO\(_2\), may be potentially important in gas-phase and heterogeneous recycling, although these species have never been directly measured. The results of this study highlight the important need for improved understanding of snow chemistry and microphysics.
4.034 Tracking the evolution of all carbon in the multigenerational oxidation of biogenic organic compounds.

Early Career Scientist

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Abstract:

Atmospheric oxidation processes quickly transform emitted organic compounds into a multiphase, chemically dynamic system of organic aerosol and gas-phase products. A complete understanding of the fate and transformations of organic carbon in the atmosphere therefore requires a time-resolved quantitative description of both gas- and particle-phase carbon, but attempts to understand the evolution of carbon through atmospheric oxidation has in nearly all cases resulted in a large fraction of “missing” or unidentified carbon. We present here results from laboratory oxidation experiments in which a large suite of state-of-the-art mass spectrometric and spectroscopic instrumentation was employed to measure organic compounds across nearly all volatilities and chemical functionalities typically observed in the atmosphere. Photochemical oxidation of common biogenic emissions was studied, spanning hours to days of simulated atmospheric aging, with a focus here on α-pinene. New chemical ionization mass spectrometric instruments allowed for the quantification of low-volatility gases formed through these processes, and characterization of their role in aerosol
growth. Through systematic variation of reaction conditions (e.g. with and without seed aerosol) coupled with measurements of low-volatility compounds, we explore the importance of measurement artifacts, especially vapor deposition to surfaces, on typical measurements and laboratory experiments. By observing all carbon through multiple generations of oxidation, we examine the transitions between particles and lower-volatility gases with unprecedented detail and place them in the context of current simplified (i.e. two-dimensional) model frameworks, providing a more complete understanding of the evolution of organics in the atmosphere as well as an updated assessment of the capabilities and limitations of current atmospheric instrumentation.
4.035 Can Viscous Liquids Provide Flexible Templates for Atmospheric Ice Nucleation? 

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Abstract:  
Formation of water ice in the atmosphere is often facilitated by a solid compound which acts as a template catalyzing ice nucleation at warmer temperatures than otherwise possible. While poorly understood, the efficiency of organic aerosols to catalyze nucleation events has been observed to vary with a number of properties including particle size, chemical structure, oxidation state, and as we have recently discovered, aerosol phase. A wide range of solid organic aerosols, including soot, polycyclic aromatic hydrocarbons long chain organics, soot, and even cellulose can act as ice nucleating particles (INP). Surprisingly, we have recently observed that certain viscous liquid are also efficient INP. In this study, we use an optical ice microscope apparatus equipped with a sealed cooling stage and CCD camera to examine contact freezing events between a droplet and organic hydrocarbons, in their fresh and oxidized forms. Samples are exposed to ozone to represent atmospheric oxidative aging. Fresh and oxidized samples are characterized using two methods, Fourier Transfer Infrared Spectroscopy with Horizontal Attenuated Total Reflectance (FTIR-HATR) and Raman Microspectroscopy. For the liquids included in this study, squalane and squalene, the temperature dependence of the viscosity was measured. Our results demonstrate that viscous liquids are effective catalyze ice nucleation at temperatures comparable to octacosane, and at temperatures only slightly colder than the aromatic compounds studied. The observed behavior is consistent with a flexible template molecular model of ice nucleation events. The atmospheric implications of our results are highly significant since they imply that a wide range of previously ignored organic aerosols may catalyze ice nucleation in the atmosphere provided they exhibit adequately high viscosities at ice freezing temperatures. Further measurements are needed to understand the role of viscosity in catalyzing heterogeneous nucleation.
On-line measurement of aerosol volatility at a regional background site in China.

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Abstract:

The coupling of a Thermal Denuder (TD) with a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) was setup in Xianghe, which is a regional background site in North China, during June - July, 2013 to on-line measure the mass concentrations and semi-volatilities of atmospheric submicron particles, including organic matter (OM), \( \text{SO}_4^{2-} \), \( \text{NO}_3^- \), \( \text{NH}_4^+ \), and \( \text{Cl}^- \). The total PM\(_1\) mass concentration measured was averagely \((47.9\pm47.3)\) mg/m\(^3\) during the campaign, with OM accounting for 38.2% of the total PM\(_1\) mass, followed by \( \text{SO}_4^{2-} \) (33.7%), \( \text{NH}_4^+ \) (13.8%), \( \text{NO}_3^- \) (12.3%), and \( \text{Cl}^- \) (2.0%). It was found that \( \text{NO}_3^- \) and \( \text{Cl}^- \) had the highest semi-volatility, with about 60% of them evaporating into the gas phase by increasing the temperature to 50 °C, while \( \text{SO}_4^{2-} \) showed the lowest semi-volatility, with almost 90% of its mass remaining in the particle phase at 50 °C. The semi-volatility of OM and \( \text{NH}_4^+ \) was at the middle level. The semi-volatility of \( \text{NO}_3^- \) was affected by the pollution level of the atmospheric submicron particles since it showed an increasing trend with the increasing of PM\(_1\) at 50 °C. The oxygen-to-carbon ration of organic aerosol was 0.47 to 0.60 by increasing the temperature from 50 °C to 200 °C. In addition, the semi-volatility of the PM\(_1\) species with vacuum aerodynamic diameters of 60-2000 nm was little size dependent. The calculation based on the high-resolution mass spectra of OM showed that \( \text{CO}_2^+ \)-containing organic species had lower semi-volatility, while \( \text{C}_4\text{H}_9^+ \)-containing organic species had higher semi-volatility. The semi-volatility of OM was found to be negatively related to its oxidation state. The quantitative result of atmospheric submicron particles’ semi-volatility is essential to the research of the physicochemical properties and pollution mechanism of particles as well as the validation of the air quality models.
Oxidative Potential Evolution of Particulate Trimethylamine during Ozonolysis.

Early Career Scientist

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Abstract:

ABSTRACT  
Amines in the atmosphere have attracted widespread attention because they contribute to the nitrogen cycle\(^1\), new particle formation\(^2-4\), and brown carbon\(^5\), as well as having their own inherent toxicity\(^6\). In this study, the ozonolysis of particulate trimethylamine (TMA), which was produced via heterogeneous uptake of TMA onto (NH\(_4\))\(_2\)SO\(_4\), NH\(_4\)HSO\(_4\), NH\(_4\)NO\(_3\) and NH\(_4\)Cl or neutralization of TMA and H\(_2\)SO\(_4\), was investigated using *in situ* attenuated total reflection Fourier transform infrared spectroscopy (ATR-FTIR) and proton transfer reaction mass spectrometry (PTR-MS). DTT assay tests were performed to assess oxidative potential changes due to O\(_3\) oxidation. Products including HCOOH, HCHO, CH\(_3\)N=CH\(_2\), (CH\(_3\))\(_2\)NCHO, CH\(_3\)NO\(_2\), CH\(_3\)N(OH)CHO, CH\(_3\)NHOH and H\(_2\)O were identified on all the substrates exposed to ppbv level of O\(_3\). Compared with ammonium and aminium salts, the oxidation potential of particulate aminium after ozone oxidation increased significantly, with a DTT loss rate that increased from 0±4.14×10\(^{-6}\) to 5.92±2.80×10\(^{-3}\) pmol·min\(^{-1}\)·μg\(^{-1}\). Our results reveal that the oxidation of particulate amines is a potential degradation pathway for amines in the atmosphere and that the oxidation products in the particle phase are associated with modification of the adverse health impacts of aerosol particles.

REFERENCES  
4.039 Heterogeneous reaction of SO2 with soot: the role of the chemical composition of soot in surface sulfates formation.

Early Career Scientist

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Abstract:

It has been found that soot can catalyze the oxidation of SO2 to form sulfates in the presence of O2 and water\textsuperscript{1, 2}. A linear relationship has been observed between NO\textsubscript{2} uptake and the content of reduced organic carbon in soot prepared under different combustion conditions\textsuperscript{3}. However, the effect of the chemical composition of soot on the adsorption or further oxidation of SO\textsubscript{2} on the surface of soot is still poorly understood. In this study, soot samples with different fractions of unsaturated hydrocarbons and oxygen groups were prepared by combusting n-hexane under well-controlled conditions. The heterogeneous reaction of SO\textsubscript{2} with soot at ambient pressure and ambient relative humidity (RH) was investigated using \textit{in situ} attenuated total internal reflection infrared (ATR-IR) spectroscopy, ion chromatography (IC) and a flow tube reactor. We observed that the surface properties of soot, which were governed by combustion conditions, played an important role in the heterogeneous reaction of SO\textsubscript{2} with soot. This role was found to greatly depend on RH. At low RH, soot produced with high fuel/oxygen ratio exhibited high reactivity toward SO\textsubscript{2}, because it contained a large amount of unsaturated hydrocarbons which acted as the active site for SO\textsubscript{2} adsorption. At moderate RH, water and SO\textsubscript{2} both participated in the reaction. Soot produced with moderate fuel/oxygen ratio showed high reactivity toward SO\textsubscript{2}, since it contained appropriate amounts of unsaturated hydrocarbons and oxygen groups, which led to optimal surface concentrations of SO\textsubscript{2} and water, respectively.

REFERENCES
4.040 Chemical Aging of Primary and Secondary Organic Aerosol.

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Abstract:
Secondary organic aerosol (SOA) formation in the atmosphere can be thought of as a succession of oxidation steps. The production of later-generation SOA via continued oxidation of the first-generation products or the primary organic aerosol components is defined as chemical aging. Our work investigates the changes in composition (oxygen to carbon ratio, average mass spectrum), physical properties (volatility, density, water solubility), and mass concentration of organic aerosol (OA) as its components continue reacting. A series of experiments was conducted in the environmental simulation chambers at Carnegie Mellon University and the University of Patras. The fresh particulate matter consisted of secondary OA (from the photooxidation of a-pinene, b-caryophyllene, toluene, xylenes), primary OA (from cooking, wood burning), and model secondary OA compounds (MBTCA). This first-generation OA was allowed to react further with hydroxyl radicals (OH) generated via HONO photo-dissociation. The SOA was characterized by a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-AMS) and the gas phase by a Proton Transfer Reaction Mass Spectrometer (PTR-MS). Additional measurements included the density of the OA, its volatility, and hygroscopicity. Positive matrix factorization was used to separate the fresh and aged OA in these experiments. A wide variety in behavior was observed during these chemical aging experiments. For some systems additional OA was formed and the oxygen to carbon increased suggesting that the functionalization pathway dominated the various reactions. For some others (mainly highly oxidized SOA), there was a decrease of the oxygen to carbon ratio and of the mass concentration indicating that the fragmentation of later-generation SOA products was the dominant process. The results for the various systems investigated are summarized and synthesized.

Early Career Scientist

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Abstract:

Organosulfates (OSs) are ubiquitous components of particulate matter in atmospheric environment. They are formed as a result of interactions of organic compounds or their oxidation products with acidic sulfate particles. Two types of ultra-high resolution mass spectrometers (UHRMS), Fourier Transform Ion Cyclotron Resonance mass spectrometer (FT-ICR MS) and Orbitrap MS, were used to characterize OSs in aerosol samples collected in the Pearl River Delta, China. With the remarkable resolving power and mass accuracy of UHRMS, hundreds of molecular formulas can be assigned to sulfur-containing compounds (CHOS- and CHONS-), of which OSs possibly make up the majority. FT-ICR and Orbitrap have different mass resolution and mass accuracy. Both have been used to characterize organic aerosols previously, but there isn’t comparison of formulas identified by the two UHRMS. In our study employing both UHRMS, 817 OS formulas were identified using FT-ICR while 304 OS formulas using Orbitrap. OS formulas of high s/n (>400) detected in FT-ICR analysis was also found in Orbitrap analysis, but for OS formulas of lower s/n, the percentage of overlapping detection by both UHRMS became increasing small with declining s/n. The sensitivity difference of the two UHRMS was the dominant cause for accounting for 99% of the discrepancy in the number of identified formulas. Another cause for formula number difference is the superior mass accuracy of FT-ICR (<1ppm) over Orbitrap (<2ppm), as some m/z peaks by Orbitrap could not be assigned a unique formulas and thus not reported previously. This comparison suggests that improved MS analysis optimization with Orbitrap, such as scanning small m/z ranges, is needed to minimize mass detection bias. Otherwise part of molecular information would
be lost. The numerous OS formulas identified in this UHRMS study also indicate the high complexity of OS precursors and/or formation pathways in the atmosphere of polluted regions in China.
Decadal and seasonal distributions of low molecular weight dicarboxylic acids and related compounds, and their stable carbon isotope ratios in the remote marine aerosols from the western North Pacific.

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Abstract:

Oxalic and other dicarboxylic acids comprise an important fraction of water-soluble organic aerosols in various environments. To better understand long-term atmospheric changes in the western North Pacific, we collected marine aerosol samples in 2001-2014 on weekly basis at a remote island, Chichijima (27°04’E; 142°13’N), which is located in the boundary of westerly and easterly wind regimes. Here we present seasonal and decadal distributions of dicarboxylic acids (C₂-C₁₁), oxoacids (C₂-C₉) and a-dicarbonyls (C₂-C₃), which were determined using gas chromatography (GC) and GC/MS techniques after BF₃/n-butanol derivatization. We also measured stable carbon isotopic composition (d¹³C) of diacids and related compounds using a GC/isotope ratio/MS technique. Concentrations of total diacids fluctuated in a range of 10-600 ng m⁻³ with winter/spring maxima and summer minima. The maximum concentrations in winter/spring can be explained by a combination of enhanced emissions of polluted aerosols and their precursors in the Asian Continent and enhanced atmospheric transport to the North Pacific due to the intensified westerly winds in winter/spring. The concentrations of diacids seemed to increase from 2001 to 2008 and then decrease toward recent years, as supported by major ion analyses. Recent decline of diacids concentrations may be associated with the changes in the anthropogenic emissions due to the air quality improvement in Asian countries.

Stable carbon isotopic compositions determined for 2006 showed relatively high d¹³C values of oxalic acid (-22‰ to -4‰). The isotopic ratios were found to increase from winter to summer. Significant enrichment of ¹³C in oxalic acid in summer should be associated with photochemical aging of organic aerosols in the marine atmosphere, caused by kinetic isotopic fractionation during the photo degradation of oxalic acid. We will discuss the decadal changes in the stable carbon composition of oxalic acid in relation to the atmospheric oxidation capability in the western North Pacific.
Abstract:

Secondary organic aerosol (SOA) may form as a result of chemical reactions in cloud droplets which are referred to as “cloud aqSOA”. Clouds can dissolve organic vapors in the aqueous phase where these compounds are transformed into SOA. Accretion and oxidation processes may compete in the aqueous phase. Cloud chemistry acts both as a source for SOA (through oligomerization and functionalization reactions) and as a sink for SOA (through fragmentation reactions).

Cloud chemistry has been developed to represent and evaluate these chemical processes and recent modeling studies aimed at implementing newly identified accretion processes to evaluate their potential impacts on SOA formation. However, a matching accurate knowledge of aqueous oxidation is required since oxidation processes may control the organic radicals availability to form “accretion” products. Due to the numerous organic compounds found in cloud water, several oxidation pathways are not fully documented. To remedy this lack of information, structure activity relationships (SAR) are now available in the literature for estimating missing parameters such as kinetic constants, branching ratios, hydration constants, acidity constants, etc. Based on these structure-activity relationships, a new detailed aqueous phase mechanism (CLEPS: CLoud Explicit Physico-chemical Scheme) describing the oxidation of soluble organic compounds resulting from isoprene oxidation is proposed.

This new aqueous phase mechanism is coupled with the detailed gas phase mechanism MCM v3.3. The GROMHE SAR allows for the evaluation of Henry's law constants for organic compounds. Variable photolysis in both gas and aqueous phases using the TUV 4.5 radiative transfer model is calculated. The resulting multiphase mechanism has been implemented in a cloud chemistry model, using a warm microphysical scheme module.

Sensitivity tests experiments are presented on oxygenated compounds produced by the isoprene oxidation.
Introducing biological processes in cloud chemistry models.

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Abstract:
Models have been developed to study the multiphase cloud chemistry. But, only abiotic processes have been considered in these models to represent the transformations of organic and inorganic species. Recently, microorganisms (bacteria, yeasts and fungi) have been discovered as new potential actors of cloud chemistry. The presence of metabolic activity in clouds leads to an uptake of molecular compounds by cells as nutrients but also to a production of larger molecules. Microorganisms are therefore considered as biological catalysts and they potentially compete with chemical reactions in cloud aqueous phase.

Microorganisms can degrade carbon compounds such as formaldehyde, methanol and organic acids thanks to their carbon metabolism. They also interact with oxidants: they consume $\text{H}_2\text{O}_2$ that is a major source of $\text{HO}^*$ radicals due to their oxidative stress metabolisms. Recently, comparisons between the rates of biotic and abiotic transformations in microcosms mimicking cloud environments have clearly demonstrated that biodegradation can be competitive with radical chemistry. However, such laboratory experiments have been performed under “bulk” conditions that reflect only aqueous phase reactivity. But the cloud system is a multiphase (gas/liquid/solid) medium where chemical processes take place and are modified by microphysical and dynamical processes. The next step is thus to integrate biological reactions within cloud chemistry models.

For this, biodegradation rates were experimentally determined for 4 chemical compounds (formic and acetic acids, formaldehyde and $\text{H}_2\text{O}_2$). Three bacteria strains isolated from the cloud water sampled at the puy de Dôme (France) were selected for their capacity to efficiently degrade these compounds. These biodegradation rates are introduced for the first time in a multiphase cloud chemistry model using the CLEPS aqueous mechanism (Cloud Explicit Physico-chemical Scheme). The objective of this work is to compare radical vs. microbiology efficiency looking at the effect of selected “key” environmental parameters such as temperature and solar irradiation intensity.
4.048 Oxidation flow reactors (OFRs) to study secondary aerosol formation: overview of recent field and modeling studies.

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Abstract:

OFRs allow studying SOA formation and aging in both laboratory and field experiments. The concentration of an oxidant (OH, O$_3$, or NO$_3$) can be increased, leading to hours–months of equivalent atmospheric oxidation during the several-minute residence time. Typically, more SOA formation is observed from nighttime than daytime air. Measured ambient VOCs cannot explain the observed SOA formation, suggesting that typically unmeasured S/IVOCs (possibly VOC oxidation products or direct emissions) are important ambient SOA precursors. A kinetic model is used to study OFR chemistry. OH exposure ($\text{OH}_{\text{exp}}$) can be estimated within a factor of ~3 using model-derived equations, and verified using VOC decay measurements. $\text{OH}_{\text{exp}}$ is strongly dependent on external OH reactivity, which may cause significant OH suppression in some circumstances (e.g., lab/source studies with high precursor concentrations). UV photolysis and O atoms are typically minor reaction pathways, except under high OH suppression. Low-volatility organic gases (LVOCs) fate is dependent on particle condensational sink. For the range of particle condensational sink at a remote pine forest, ~60% of produced LVOCs were predicted to condense onto aerosols for an OH$_{\text{exp}}$ of ~1 day, with the remainder lost to walls. Similar to chamber wall loss corrections, a correction is needed to relate OFR sampling to the atmosphere, where condensation onto aerosols is the dominant LVOC fate. At high OH$_{\text{exp}}$ (>20 days) in an OFR, LVOCs are predicted to be oxidized many times, leading to formation of volatile fragmentation products that no longer form SOA. Changes to preexisting OA at high OH$_{\text{exp}}$ are a result of heterogeneous oxidation. The fate of RO$_2$ and NO$_3$ is being investigated with the model. SOA yields specific to OFR oxidation were investigated using standard addition of individual VOCs into ambient air, and were consistent with laboratory large chamber yields.
**4.049 Evaluation of updated isoprene, terpene and aromatic oxidation in the MOZART chemical mechanism.**

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**Abstract:**

Recent laboratory and field observations have provided important advances in our understanding of the oxidation pathways of isoprene and terpenes, which are the major natural contributors of volatile organic compounds (VOCs) to the atmosphere, and precursors to ozone and secondary organic aerosols (SOA). Intermediate isoprene oxidation products, such as isoprene epoxydiols (IEPOX), hydroperoxy aldehydes (HPALD), isoprene peroxides and isoprene nitrates, have been measured during several field campaigns in the Southeast United States. The MOZART tropospheric chemistry mechanism has been expanded to specifically represent these measured intermediate compounds, thus allowing for quantitative evaluation of the model representation of the emissions and chemical evolution of isoprene and terpenes. The MOZART chemistry has also been expanded to treat specific aromatic species (benzene, toluene, xylenes) instead of a single lumped aromatic compound. The NSF/NCAR/NASA DC3, NSF/NCAR NOMADSS, NASA SEAC4RS and NSF/NCAR FRAPPE aircraft campaigns provide simultaneous observations of numerous VOCs using several instruments and measurement techniques, such as TOGA (Trace Organic Gas Analyzer, fast GC-MS), WAS (whole air sampler), CIMS (chemical ionization mass spectroscopy) and PTR-MS (proton-transfer-reaction mass spectroscopy). The MOZART chemical mechanism is used in both a box model (BOXMOX) and the global chemistry climate model CAM-chem (Community Atmosphere Model with Chemistry, a component of the Community Earth System Model) for evaluation and comparison to previous chemistry schemes. BOXMOX is initialized using the field campaign observations for a variety of locations to evaluate the chemistry under a mixture of natural and urban conditions. CAM-chem is run with specified dynamics to represent the specific dates of the campaigns and allow direct comparison between model results and observations. CAM-chem includes online biogenic emissions using the MEGAN v2.1 algorithms, and the airborne measurements are used to evaluate
the biogenic emissions of isoprene, monoterpenes, methanol, acetone, acetaldehyde, and other compounds.
A global modelling study of the release of ClNO2 from tropospheric aerosol and its impact on tropospheric oxidation.

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Abstract:
Nitrogen oxides play a central role in the chemistry of the atmosphere, affecting levels of both ozone and OH. Heterogeneous removal of the NOx reservoir, N2O5, onto aerosol particles can be a major loss route for NOx with modelling work by Tie et al. (2003) suggesting that, at high latitudes, N2O5 hydrolysis can reduce NOx levels by as much as 90%. The reactivity of the aerosol towards N2O5 has been shown to be a complex function of ambient temperature and RH as well as aerosol composition. Following measurements by Osthoff (2008), Thornton and co-workers demonstrated that the presence of chloride ions in the aerosol can release nitryl chloride, ClNO2, following uptake of N2O5. The night-time chemistry leads to a build-up of nitryl chloride, which can subsequently be photolysed to yield chlorine radicals, an atmospheric oxidant, and NO2, regenerating NOx. The yield of ClNO2 depends on particulate levels of chloride and nitrate, as well as factors controlling initial N2O5 uptake. The production of ClNO2 at the air-sea interface has been studied by Kim et al., and the important role of surface active organics demonstrated by Ryder et al.

We have included these processes in a chemistry-climate mode, the UK Met Office Unified Model, UM/UKCA-MODE, using a parameterised yield of ClNO2 from N2O5 aerosol uptake. In this paper, the performance of the parameterisation is examined and the effect of the halogen chemistry on levels of e.g. ozone and particulate nitrate will be investigated. Comparison with field measurements e.g. Tang et al. will also be made, and the impact of ClNO2 release on oxidative chemistry in the troposphere quantified.
4.052 OH reactivity and experimental OH budget in Wangdu (North China Plane) in summer 2014.

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Abstract:

OH reactivity was measured via pump and probe technique and direct detection of OH radicals by laser-induced fluorescence during a four week-long campaign in Wangdu in summer 2014. Wangdu is a small city located approximately 170 km south-west from Beijing in the North China Plane. A large set of instruments measured together to characterize regional photochemistry. Gas-phase measurements included species to experimentally determine the production and destruction rates of OH radicals and inorganic and organic species to compare direct measured OH reactivity with the sum of reactivity from single species. OH reactivity median values ranged between 10 and 20 1/s during daytime with slightly higher values during the first part of the campaign. Overall, contributions of measured single compounds to the total OH reactivity can explain nearly 80% of the measured OH reactivity. During the second part of the campaign, the entire measured OH reactivity is explained by measured concentrations of OH reactants. Similarly, the sum of OH production rates calculated from measurements is balanced by the OH destruction rate during this time. In contrast, the OH destruction rate is approximately 20-30% larger than the sum of OH production rates during noontime and in the afternoon during the first part of the campaign.
4.053 Interaction of acidic trace gases with ice: XPS and NEXAFS analysis with the new NAPP chamber for solid interfaces at SLS.

Early Career Scientist

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Abstract:

Acidic trace gases, such as HCl and formic acid, play an important role in atmospheric chemistry. The presence of ice disturbs this chemistry. The molecular processes of the interaction of acidic trace gases with ice are still a matter of debate. Neither a quantification of the uptake is possible nor a molecular picture of the uptake is available at temperatures relevant to Earth’s snow cover (Bartels-Rausch et al. 2014 (Atmos. Chem. Phys.) Huthwelker et al. 2006 (Chem. Rev.)). This hinders the development of global models that would allow predicting changes to the large-scale effects of ice clouds. So far, direct experimental observations of the ice surface and of the interaction with trace gases at temperatures and concentrations relevant to the environment are limited. In this study, we take advantage of the surface and analytical sensitivity as well as chemical selectivity of photoemission and absorption spectroscopy performed at ambient to overcome this limitation in environmental science. The usage of X-ray Photoelectron Spectroscopy (XPS) allows us to get the concentration depth profile of dopants. To perform measurements close to atmospheric conditions the Near-Ambient-Pressure-Photospectroscopy endstation (NAPP) chamber at SLS was optimized (Orlando et al. 2016 (Topics in Catalysis)). With the combination of XPS and Near-Edge X-ray Absorption Finestructure (NEXAFS) we get information whether the interaction with the trace gas leads to enhanced surface disorder of the ice surface and to what extent this change influences the uptake of the trace gas.

Here we present the first analysis of the interaction and uptake of HCOOH with ice by means of electron spectroscopy. Depth profiles indicate that the dosed acid stays at the ice surface as long as disorder is not enhanced. The results are compared to earlier
analysis using different approaches and trace gases.
### 4.055 Importance of reactive halogens in the tropical marine atmosphere.

**Early Career Scientist**

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**Abstract:**

Halogen species (chlorine, bromine and iodine) are known to play an important role in the chemistry and oxidizing capacity of the troposphere, particularly in the marine boundary layer (MBL). Reactive halogens cause ozone (O$_3$) destruction, change the HO$_X$ and NO$_X$ partitioning, affect the oxidation of volatile organic compounds (VOCs) and mercury, reduce the lifetime of methane, and take part in new particle formation.

Numerical models predicted that reactive halogen compounds account for 30% of O$_3$ destruction in the MBL and 5-20% globally. Up to 34% of O$_3$ loss is due to I and Br combined in the TORERO area.

Recent studies have highlighted the key role that heterogeneous chemistry plays in explaining observations of BrO and IO abundances in the tropical troposphere. Moreover, it has been suggested that halogen species undergo heterogeneous reactions on ice surfaces.

The main objective of this study is to investigate the atmospheric chemistry in the tropical East Pacific with a focus on reactive halogens using the latest version of the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) and field data from the TORERO campaign. WRF-Chem is a highly flexible community model for atmospheric research where aerosol-radiation-cloud feedback processes are taken into account. Our reaction mechanism in WRF-Chem is based on the MOZART mechanism and has been extended to include bromine, chlorine and iodine chemistry. Heterogeneous recycling reactions involving sea-salt aerosol and ice particles have been included into
the model, along with oceanic emissions of important OVOCs and halocarbons. Sea surface emissions of inorganic iodine are calculated using the parameterisation of Carpenter et al. (2013). Focusing on TORERO observations from the ships and a selected number of flights we present an evaluation of the relevant tropospheric gas-phase chemistry (O₃, H₂O), inorganic halogen species (BrO, IO), aldehydes (CH₃CHO, CHOCHO) and Very Short Lived Halocarbons (VSLH).
Abstract:

Atmospheric chemistry fundamentals are heavily fed by laboratory kinetic and photochemical studies. In the TROPOS ACD aqueous phase chemistry laboratory, such reactions are investigated systematically. Hence, in the first part of the presentation, recent results from these fundamental laboratory studies will be presented and discussed. Systems studied include: (i) Peroxyl radical formation - knowns and unknowns, (ii) the photochemistry of iron-dicarboxylate complexes and (iii) small inorganic radical chemistry - an approach for studying reactions under high electrolyte concentrations. Fundamental studies must be linked to the real atmosphere. In the second part the so-called Hill Cap Cloud Thuringia 2010 (HCCT-2010) experiment is outlined which has been performed with a number of collaborating groups to study aerosol-cloud interaction with an emphasis on chemical processing in particles and clouds. Selected results of this campaign will be presented and discussed. These results relate to the occurrence of functionalised organic compounds in cloudwater, mass closure and in-cloud processing as well as effects of cloud on gas phase HOx.

Finally, a few thoughts will be presented on how fundamental studies can be better embedded into atmospheric chemistry research and an outlook on the further development of multiphase chemistry as an active field within atmospheric chemistry will be discussed.
Long-term variation of precipitation frequency and its correlations with aerosols in China during the past 50 years.

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Abstract:

The spatial and temporal variations of the global precipitation have been investigated both in the frequency and the amount since the past several decades. Confident evidence have shown the significant increasing of the heavy rainfall but decreasing of the light one both in observation and multi-model simulations. Most studies connected it to the global warming, which brings the unbalance between the increasing magnitude of global evaporation (2-3% K⁻¹) and precipitation intensity (7% K⁻¹) according to Clusius-Clapeyron equation, and indicated the decreasing of light precipitation. And a number of studies suggested the effects of the aerosols are responsible for the variation of global precipitation via changing the cloud microphysical properties and solar radiation balance. In this study, 50 years (1964 - 2013) monthly averaged precipitation frequency (PF), evaporation, cloud fraction data from the Climatic Research Unit (CRU) with the resolution of 0.5°×0.5°, as well as the daily precipitation amount, surface temperature and visibility data at more than 2000 observation sites from China Meteorological Administration (CMA) were used to analyze the spatial distributions and the temporal variations of precipitation in China based on the Combined Empirical orthogonal functions (CEOF) method. The significant changes of the PF distribution were located in Beijing-Tianjin-Hebei (BTH) and Inner Mongolia, where sandstorm and heavy haze occurred frequently, indicating a strong link might exists between the aerosols and PF. Qiu’s parameterization model of aerosol optical depths (AOD) in China inversed based on the observed visibility was deployed to characterize the aerosol concentrations. A good correlation of the time-series of the first EOF eigenvector was found between the AOD and PF. Although there is a great challenge on determining which dominants the variations of precipitation in China, further study might focus more on the variation of PF rather than the amount.
4.058 Molecular Composition of Biomass Burning Aerosol from Household Cookstoves in Rural Haryana, India.

Early Career Scientist

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Abstract:

Exposure to PM2.5 is associated with cardiopulmonary and allergic diseases, and it is known to increase mortality and morbidity in the general population. Emissions of air pollutants, such as PM2.5, from biomass-burning cookstoves in India have proven to be significant on regional scales. However, the extent of competition between the primary particle evaporation, which reduces the PM2.5 mass, and chemical oxidation of VOC components in smoke, which increases the PM2.5 mass by forming secondary pollutants, is poorly understood. To better constrain the contribution of primary and secondary aerosols to the aged cookstove smoke, we collected particulate matter and whole air samples from prescribed cooking activities using traditional cookstoves in Palwal District, Haryana, India. In addition, aerosol samples were collected under controlled cooking conditions in a village home. The fuel sources included brushwood and dung. Moisture content of the fuel was controlled for; other variables included the stove type and meal cooked. The whole air samples were analyzed using gas chromatography methods to determine the distribution of non-methane volatile organic compounds (NMVOCs) in fresh smoke. Dung fuel resulted in a significantly lower modified combustion efficiency with simultaneously larger emissions of measured VOCs. The molecular composition of the particulate organic compounds was characterized using nanospray desorption electrospray ionization (nano-DESI) high-resolution mass spectrometry. Nitrogen-containing compounds dominate the nano-DESI-MS signal in dung smoke samples, while the majority of detected compounds from brushwood smoke do not contain nitrogen. Both levoglucosan and potassium biomass-burning tracers were abundantly detected in brushwood samples, while only some dung smoke samples contained levoglucosan. The results of this study will be used as an input to a model that describes secondary chemical processing of smoke, and will serve as basis for determining the effect of replacement of traditional cookstoves with more modern alternatives.
4.059 Secondary Organic Aerosol Formation from the Green Leaf Volatile Cis-3-Hexen-1-ol.

Early Career Scientist

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Abstract:

Cis-3-hexen-1-ol (HXO) is a type of green leaf volatile, which is an important class of biogenic volatile compounds that are emitted due to plant stress. Chamber experiments were performed to examine secondary organic aerosol (SOA) formation from the photooxidation and ozonolysis of HXO using either nonacidified or acidified sulfate seed aerosol and under wet (relative humidity (RH) = 45-60%) or dry conditions (RH<6%). For select ozonolysis experiments, a hydroxyl radical (OH) scavenger was utilized. Aerosol sizing measurements and off-line chemical analyses by gas chromatography/mass spectrometry (GC/MS) and ultra performance liquid chromatography/ electrospray ionization high-resolution quadrupole time-of-flight mass spectrometry (UPLC/ESI-HR-Q-TOFMS) were used to characterize HXO-derived SOA formation. Our results indicate that the level of HXO-derived SOA increases with elevated aerosol acidity and under low-RH conditions, indicating that the water dilutes the acidity, thus decreasing the SOA. Experiments performed in the presence of an OH scavenger showed that the SOA formation decreased compared to experiments without an OH scavenger, suggesting that the major channel contributing to SOA formation is the reaction with OH radicals. Chemical characterization of HXO-derived SOA reveals that organosulfates (OSs) were produced. Ambient fine aerosol (PM$_{2.5}$) samples collected from Rio de Janeiro, Brazil, were also analyzed. Many of the HXO-derived OSs identified in the chamber experiments
were also found in the PM$_{2.5}$ samples collected in Rio de Janeiro, Brazil. This study provides direct evidence that the atmospheric oxidation of HXO yields biogenic SOA through the formation of polar OSs.
4.060 Atmospheric Chemistry of E- and Z-CF3CH=CHCF3.

Early Career Scientist

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Abstract:

FTIR smog chamber experiments were performed to investigate the atmospheric fate of E- and Z-CF3CH=CHCF3 (1,1,1,4,4,4-hexafluoro-2-butene). The experiments were performed to study reactions of E-CF3CH=CHCF3 or Z-CF3CH=CHCF3 with Cl atoms, OH radicals, and O3 in 700 Torr of N2/O2 diluents at 296 ± 2 K. The study determined the Cl atom, OH radical, and O3 kinetics and the mechanism of the atmospheric oxidation of E- and Z-CF3CH=CHCF3. The main atmospheric fate for both compounds is reaction with OH radicals. Atmospheric chemistry of the reaction of Z-CF3CH=CHCF3 with OH and OD radicals were investigated by Baasandorj et al. in 2011 [1], no other previous studies are available. The results of the present study are compared to the findings of Baasandorj et al. assessing the atmospheric impact of E- and Z-CF3CH=CHCF3. Infrared spectra were recorded, and the atmospheric lifetimes, the radiative forcings and global warming potentials (GWP) of the two butenes were calculated. This study provides a comprehensive description of the atmospheric fate of E- and Z-CF3CH=CHCF3. We present here the first results on the atmospheric chemistry of E-CF3CH=CHCF3 and of the Cl- and O3-initiated chemistry of Z-CF3CH=CHCF3.

References

Simultaneous and time-resolved measurement of alpha-pinene ozonolysis products in gaseous and aerosol phases for the determination of gas-aerosol partitioning.

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Abstract:

We have developed a real-time measurement system to identify semi-volatile organic compounds (SVOCs) in both the gaseous and the aerosol phases using chemical ionization-mass spectrometry in order to estimate the gas-aerosol partitioning of each SVOC. In our system, SVOCs in both the gaseous and the aerosol phases are measured by the same technique, which enable us to determine the partitioning of each SVOC from the ratio of ion signals, without knowing the concentration of each SVOC. This ability is a strong point of this approach, because most chemical species in SOAs are thought to be multifunctional, and determining their concentrations seems to be impossible. We show a novel system equipped with dual-proton transfer reaction-mass spectrometers for “real-time” measurements of the partitioning of each SVOC produced in alpha-pinene ozonolysis between the gaseous and the aerosol phases. Time profiles of the SVOCs in both the gaseous and the aerosol phases, *i.e.* “real-time” SVOC partitioning, are also discussed.
Organic aerosol hygroscopicity and the contribution to CCN concentrations over a mid-latitude forest facing the North Pacific.

Early Career Scientist

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Abstract:
Biogenic secondary organic aerosol (BSOA) can be formed by the reactions of volatile organic precursors emitted from vegetation in forest environments. The formation of BSOA is potentially important to that of cloud droplets by changing the hygroscopicity and other properties of aerosols. In this study, the CCN activation of submicron aerosols and their chemical composition and size distributions were measured at a mid-latitude forest site in Japan in summer and early autumn, to assess the influence of biogenic SOA on CCN in the forest environment. The hygroscopicity of organic aerosols represented by a single parameter $k_{\text{org}}$ was determined from the CCN activation diameters and composition of the aerosols. The $k_{\text{org}}$ of organics in <100 nm particles was on average substantially lower than that of organics in >100 nm particles and $k_{\text{org}}$ related positively to the O to C atomic ratio. Histogram analysis showed that the hygroscopicity of organics explains more than 60% of total aerosol hygroscopicity for 12% and 35%-41% of the measured accumulation mode and Aitken mode particles, respectively. Based on size-resolved chemical composition measurement, the contributions of organic aerosols to total measured CCN concentrations were calculated with an alternative method assuming the absence of organics. In the case that the volume fraction of organic aerosol was larger than 80% the calculated average contributions were 50%, 53%, 66%, and 80% under supersaturations of 0.80%, 0.42%, 0.23%, and 0.11%, respectively. Further, histogram analysis indicated that organic aerosols contributed more than 50% of the total CCN concentrations during 10%-15% of the measurement periods.

Early Career Scientist

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Abstract:
Extremely low-volatility organic compounds, or ELVOC, are believed to play a crucial role in the formation of particles in the atmosphere (Ehn et al., 2014). In recent years, high molecular weight dimer esters originating from the oxidation of the biogenic volatile organic compound (VOC) α-pinene have been observed in both laboratory-generated and ambient secondary organic aerosol (SOA) (Kristensen et al., 2013). The estimated vapour pressures of the dimer esters classify these as ELVOC, and as recent studies suggest that dimer esters form rapidly through gas-phase reactions, we believe these compounds to be likely candidates involved in new particle formation from oxidation of α-pinene. Furthermore, research shows that the dimer esters only form in the presence of ozone, thus may be used as tracers for ozone-initiated oxidation of α-pinene, and are therefore indicative of enhanced anthropogenic activities.

In the newly constructed cold-room smog chamber at Aarhus University we examine the effects of sub-zero temperatures on the formation of SOA, including dimer esters, from α-pinene oxidation. This state-of-the-art Teflon chamber allows for atmospheric simulations of the oxidation α-pinene and subsequent SOA formation at temperatures down to -16 °C. In experiments involving ozonolysis of α-pinene we observe significantly (up to >4 times) higher particle formation at sub-zero temperature. Furthermore, chemical analysis of the formed particles using liquid chromatography coupled to quadrupole time-of-flight mass spectrometry shows significant differences in the chemical composition related to the experiment temperature. In particular, the concentration of the high molecular weight dimer esters showed to be highly affected. Through this work, we provide new insight into the formation of ELVOC from the oxidation of VOCs and show how changing temperatures may affect not only the formation but also the chemical composition of atmospheric particles.

Abstract:

Heterogeneous reactions between trace gases and atmospheric aerosols impact the oxidative capacity of the atmosphere, regional air quality, and climate. Despite the atmospheric significance of gas/particle interactions, these reactions remain poorly constrained in atmospheric models due to a lack of understanding of how the physicochemical properties of atmospheric particles impact reaction rates. Using N\textsubscript{2}O\textsubscript{5} as a benchmark reactive gas, we explore the role of organic coatings and particle size on the reactive uptake of N\textsubscript{2}O\textsubscript{5}. Compared to the reactive uptake of N\textsubscript{2}O\textsubscript{5} on pure ammonium bisulfate particles, particles composed of mixtures of ammonium bisulfate and organic compounds exhibited highly variable trends in the reactive uptake of N\textsubscript{2}O\textsubscript{5}, which was found to depend on particle phase and the oxygen:carbon ratio (O:C) of the organic material. Organic compounds that significantly inhibited the reactive uptake of N\textsubscript{2}O\textsubscript{5} were found to lower the diffusivity and/or solubility of N\textsubscript{2}O\textsubscript{5} in the particle. Inorganic particle composition was found to impact the length that N\textsubscript{2}O\textsubscript{5} travels within a particle before reacting, referred to as the reacto-diffusive length (l). The reactive uptake of N\textsubscript{2}O\textsubscript{5} for ammonium sulfates exhibited a size-dependency with uptake coefficients ranging from 0.016 ± 0.005 to 0.036 ± 0.001 as the surface-area weighted particle radius increased from 39 to 127 nm. In contrast, the reactive uptake of N\textsubscript{2}O\textsubscript{5} on sodium chloride particles was independent of particle size, suggesting that N\textsubscript{2}O\textsubscript{5} reacts near the particle surface. Differences in the reactivity of the N\textsubscript{2}O\textsubscript{5} intermediate, NO\textsuperscript{2+}, with water and chloride can explain the dependencies of the reactive uptake of N\textsubscript{2}O\textsubscript{5} on particle size. These results suggest that parameterizations should factor in the size-resolved composition of ambient aerosols in order to accurately assess the impact of heterogeneous reactions on air quality and climate.
4.066 Microphysical and chemical processes affecting wet removal of soluble trace gases in deep convection observed over the central U.S..

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Abstract:

We examine wet scavenging of soluble trace gases in storms observed during the Deep Convective Clouds and Chemistry (DC3) field campaign. We perform high-resolution simulations ($dx = 600m-3km$) with the Weather Research and Forecasting model with Chemistry (WRF-Chem) of a severe storm in Oklahoma, a discrete ordinary convective storm in Alabama, and a mesoscale convective system over Arkansas/Missouri/Illinois/Mississippi. Sensitivity simulations are conducted varying the fractions of soluble gases retained in ice ($r_f$) when liquid water is transformed into frozen precipitation. Scavenging efficiencies (SEs) are calculated from the model and aircraft observations. A significantly higher SE for CH$_2$O is seen for the Alabama than Oklahoma.
storms. On the other hand, significantly more H$_2$O$_2$ is scavenged in the MCS than in the Oklahoma storm, which removes more H$_2$O$_2$ than the Alabama storm. We show that aqueous chemical sinks are not significant relative to the amount of scavenging of CH$_2$O and H$_2$O$_2$ in these three storms, and therefore do not explain the observed differences in SEs. We then explore differences among storms in precipitation production, temperature-dependent solubilities of the gases, and entrainment of background air as possible explanations for the observed differences in SEs. Finally, higher $r_f$ values for CH$_2$O, CH$_3$OOH, H$_2$O$_2$ are required in the simulations of the Alabama storm and MCS than of the Oklahoma storm to produce SEs consistent with observations. To determine whether these higher $r_f$ values are necessary to compensate for model errors in dynamics or microphysics that affect SEs, we evaluate the simulated vertical velocity and graupel mass and volume fields with those retrieved from polarimetric radar. Improved understanding of processes affecting net deep convective transport of CH$_2$O and peroxides is important to predicting the production of ozone (O$_3$) in the upper troposphere, where ozone affects radiative forcing and downwind air quality.
4.068 PROFILES OF NOX AND O3 IN AN AMAZONIAN RAINFOREST: COMPARISON OF MEASURED PROFILES WITH A MULTI-LAYER CANOPY CHEMICAL EXCHANGE MODEL.

Early Career Scientist

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Abstract:

In 2011, an 80 m high walk up tower for atmospheric research was erected at the ATTO (Amazon Tall Tower Observatory) site (02°08'38.8''S, 58°59'59.5''W) in the remote Amazonian rainforest. The nearly pristine environment allows biosphere-atmosphere studies within an ecosystem far away from large anthropogenic emission sources. Since April 2012 vertical mixing ratio profiles of H$_2$O, CO$_2$ and O$_3$ were measured at 8 different heights between 0.05 m and 79.3 m. During five intensive campaigns (Oct-Dec 2012, Oct-Nov 2013, Mar 2014, Aug-Sep 2014, Oct-Dec 2015) nitric oxide (NO) and nitrogen dioxide (NO$_2$) were also measured. Since the end of 2015 NO$_X$ measurements are performed continuously.

We applied the Multi-layer Canopy Chemical Exchange Model – MLC-CHEM to support the analysis of the observed profiles of NO$_X$ and O$_3$. This includes inferring bi-directional surface-atmosphere exchange fluxes as well as the role of the canopy interactions between the emissions, dry deposition, chemistry and turbulent transport of trace gases. During our investigation of diurnal and seasonal differences between model and
measurements, we conducted a set of sensitivity studies to analyse the effects of changes in NO\textsubscript{x}-soil emissions, in-canopy turbulence and resistances for O\textsubscript{3} and NO\textsubscript{2} uptake on wet surfaces. These analyses suggest some modification in the representation of some of the poorly constrained canopy processes resulting in a significantly better agreement between the simulated and measured exchange fluxes and concentrations. Furthermore we have compared different NO\textsubscript{x}-soil emission flux-scenarios with NO\textsubscript{x}-fluxes measured by different techniques.
4.069 Effects of Ammonia on SOA Formation and Composition.

Early Career Scientist

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Abstract:

Particulate matter (PM) is comprised of suspended particles in the atmosphere large enough to diminish visibility or affect temperature by absorbing or scattering light. Secondary organic aerosols (SOA), a major component of PM, are largely formed from the oxidation of volatile organic compounds (VOC). Despite research efforts to understand SOA formation from VOC reactions with oxidants such as OH, O₃, and NOₓ, a large uncertainty remains on how ammonia (NH₃) affects such reactions. NH₃ is widely released from agriculture and other natural and industrial sources. The U.S. Department of Commerce estimates the U.S. produces 9 million metric tons of NH₃ annually rendering it among the top 5 producing countries. This study investigates the effects of ammonia on SOA formation, optical properties, and chemical composition. SOA is formed from the oxidation of anthropogenic- and biogenic-relevant VOCs in a smog chamber at the desired relative humidity. After SOA formation, NH₃ is introduced into the chamber. The particle growth is monitored with a scanning mobility particle sizer (SMPS). A Proton-Transfer-Reaction Time-of-Flight Mass Spectrometer (PTR-ToF-MS) is used to track VOCs and a Time-of-Flight Aerosol Mass Spectrometer (ToF-AMS) is used to analyze particle composition. Subsequently, samples are collected and analyzed via direct analysis in real time mass spectrometry (DART-MS) and electrospray ionization mass spectrometry (ESI-MS). These techniques are used to determine whether NH₃ reacts with SOA to form nitrogen-containing compounds. Additionally, absorption coefficient of SOA extracts is measured. The experimental results from this study will be incorporated into two models: 1) UCI-CIT, an airshed model that contains comprehensive SOA chemistry for the South Coast Air Basin of California and 2) a coupled meteorological-air quality model for continental-scale modeling of the U.S. The combination of experimental and modeling results will allow us to evaluate the impact of NH₃ on SOA and ultimately air quality and climate.
4.071 Quantifying the sources of atmospheric ice nucleating particles from prescribed burns and wildfires.

Early Career Scientist

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Abstract:

Despite being a basic atmospheric process, our current understanding of ice nucleation in the atmosphere is low. One reason for this low understanding is that ice nucleating particle (INP) concentrations are low (only \(~1 \times 10^5\) particles in the free troposphere nucleate ice), making it challenging to identify both the composition and sources of ambient INP. Carbonaceous combustion aerosol produced from biomass burning is one potential source of INP, as it can contribute to over one-third of all aerosol in the North American free troposphere. Unfortunately, previous results from both field measurements and laboratory studies are in conflict, with estimates of the impact of carbonaceous combustion aerosol ranging from no impact to rivaling the well-known INP mineral dust. It is, however, becoming clear that the ice nucleation activity of these aerosols depend greatly on both their fuel type and combustion conditions. Given these dependencies, we propose that sampling from real-world biomass burning sources would provide the most useful new information on the contribution of carbonaceous combustion aerosols to atmospheric INP. In this work, we will present recent results looking at the sources of INP from prescribed burns and wildfires. To determine the specific contribution of refractory black carbon (rBC) to INP concentrations, we have coupled the Single Particle Soot Photometer (SP2) to the Colorado State University Continuous Flow Diffusion Chamber (CFDC). The SP2 utilizes laser-induced incandescence to quantify rBC mass on a particle-by-particle basis; in doing so, it also selectively destroys rBC particles by heating them to their vaporization temperature. Thus, the SP2 can be used as a selective pre-filter for rBC into the CFDC. Furthermore, we have also use a filter-based technique for measuring INP, the Ice Spectrometer, which can use pretreatments such as heating and digestion by H_2O_2 to determine the contribution of biological and organic particles, respectively.
Gas and particle phase characterization of limonene ozonolysis products and their role in SOA formation.

Early Career Scientist

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Abstract:

Limonene is a volatile organic compound (VOC) emitted by vegetation and can be found as a common ingredient in many household products. It exhibits an interesting chemistry owing to its two very different double bonds: a trisubstituted endocyclic and a terminal exocyclic double bond. Both double bonds can be oxidised by ozone with little or no carbon loss. The resulting products will be polar, of lower volatility and subsequently form secondary organic aerosol (SOA). SOA influences the climate and poses a risk on human health. The ozonolysis of limonene produces significant amounts of carboxylic acids which contribute to SOA formation owing to their low vapor pressures. Detailed gas phase mechanisms and information on limonene oxidation products are available through the master chemical mechanism (MCM) and form a framework against which observed product distributions can be compared.

I will present recent lab measurements of limonene ozonolysis products generated in our G-FROST facility and comparisons that have been made with numerical model calculations containing a detailed chemical mechanism (MCM). The measurement of the oxidation products in gas and particle phase was performed with an acetate High-Resolution Time-of-Flight Chemical Ionization Mass Spectrometer (HR-ToF-CIMS) coupled to a Filter Inlet for Gases and AEROsols (FIGAERO).

Early Career Scientist

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Abstract:
Methyl-buten-ol (MBO) molecules are emitted from many different natural sources. The most important biogenic emitted MBO is the 2-methyl-3-buten-2-ol (MBO232), with an estimated global emission around 1.6-2.2 Tg yr\textsuperscript{-1}. The oxidation of MBO232 produces secondary organic aerosols (SOA), when the NO concentration is low.

We investigate the possible hydrogen shift (H-shift) reactions in the peroxy radicals derived from four different MBOs; 2-Methyl-3-buten-2-ol (MBO232), 2-Methyl-3-buten-1-ol (MBO231), 3-Methyl-3-buten-2-ol (MBO332) and 3-Methyl-3-buten-1-ol (MBO331), with quantum mechanical calculations. The rate constants of the forward 1,5 H-shift reactions in all four MBO peroxy radicals are greater than the rate constant of the forward 1,4 or 1,6 H-shift reactions. The rate constants for the 1,5 H-shift reaction from a CH group or OH group are approximately 1 s\textsuperscript{-1} and 10\textsuperscript{-3} s\textsuperscript{-1}, respectively. The atmospheric impact of OH oxidation of MBO232 is investigated. The major atmospheric reactions of the MBO232 peroxy radical are the reactions with NO and HO\textsubscript{2}, with reaction yield of 84 % and 12 %, respectively. The H-shift reactions of MBO232 peroxy radical play a minor role with a total yield of about 4 %. 
Online measurements and modeling of SOA formation from isoprene photooxidation: insights from multiple chamber experiments and field campaigns.

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Abstract:

Isoprene, the most abundantly emitted non-methane volatile organic compound globally, has the potential to produce large quantities of secondary organic aerosol (SOA) with implications for climate, air quality and human health. However, much remains unknown about the mechanisms and processes that lead to isoprene derived SOA, as well as its chemical and physical properties. We present measurements and modeling of a suite of newly detected compounds from isoprene oxidation during laboratory studies at the Pacific Northwest National Laboratory (PNNL) as well as in the atmosphere during the Southern Oxidant and Aerosol Study (SOAS) field campaign. In both cases, measurements were made with a high resolution time of flight chemical ionization mass spectrometer utilizing iodide adduct ionization coupled to the Filter Inlet for Gas and AEROsol (FIGAERO) for the simultaneous sampling of the gas and aerosol phases. In chamber experiments, isoprene photo-oxidation with dry neutral seed, and, separately isoprene-derived epoxy diol (IEPOX) multiphase chemistry on aqueous particles, were investigated at a variety of atmospherically relevant conditions. Isoprene photo-oxidation under high HO$_2$ and low NO conditions produced unexpectedly large amounts of SOA, at a yield similar in magnitude but from a distinctly different mechanism than that from IEPOX uptake. Aging experiments in both cases show clear evidence for particle phase chemistry such as hydrolysis and oligomerization altering the chemical and physical properties of the SOA. Indeed, oligomerization was inferred by observations of composition and thermal desorption characteristics to be a dominant feature in NOx-rich isoprene photochemical SOA formation, although at lower overall SOA yields. Using observations and a new MCM-based gas-particle partitioning model, we assess the degree to which the different mechanisms are operable in the atmosphere and other independent chamber experiments.
Towards a complete picture of the atmospheric radical cycles: the speciated detection of gas-phase organic radicals by proton transfer mass spectrometry.

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Abstract:

The understanding of the atmospheric radical cycles and of atmosphere's oxidative capacity is mostly limited today by technical challenges: the inability of current techniques to monitor specific organic radicals in the atmosphere ("speciated detection"), especially organic peroxo radicals ("RO₂", where R is an organic group) that are intermediates in the atmospheric oxidation of most organic gases. Past and current techniques employed to monitor these radicals in the atmosphere (PERCA, ROxMax, PerCIMS, ROXLIF...) convert them into a single species (NO₂, H₂SO₄ or HO₂/OH) and provide thus overall concentrations or, at best, semi-speciated ones, distinguishing between saturated and unsaturated radicals. While valuable, this information is not sufficient for a full picture of the radical cycles.

Over the last decades, mass spectrometry using chemical ionization has been explored for its ability to detect different organic radical separately (fully speciated detection). Ionization by proton transfer was found to be the most promising approach and instruments based on this principle were built and applied to the detection gas-phase organic radicals. The first applications of this technique will be presented, as well as the many groundbreaking possibilities it offers for the understanding of the radical cycles, such as the investigation of reactions involving several organic peroxo radicals (cross-reactions, radical cycling ...), of their uptake by atmospheric surfaces, of other types of organic radicals and, ultimately, the speciated detection of organic radicals directly in the atmosphere.
Abstract:

The total reactivity of hydroxyl radical (OH) is an important tool to assess the exhaustiveness of measurements of individual compounds during intensive measurement campaigns and while monitoring air chemical composition at atmospheric stations. This approach was initiated in the early 2000s (Kovacs and Brune, 2001), based on laser induced fluorescence (LIF) techniques. Shortly thereafter, a more affordable indirect method to measure OH reactivity has been developed, namely the Comparative Reactivity Method (CRM, Sinha et al., 2008). It relies on competition reactions for OH between a reference compound with known reaction rate added to ambient air and does not make use of lasers.

Our group started recently to develop a CRM system and it has been deployed from late January to end of February 2016 close to the main SMEAR III site (third Station for Measuring Ecosystem-Atmosphere Relations), a semi-urban station in Helsinki, Finland. The main objective was to assess the CRM performances, but the results also show insight into winter OH reactivity from this semi-urban site, covering various meteorological conditions with ambient temperatures ranging from -7 to 4°C. In addition to total OH reactivity measurements reactive organic compounds were also measured with an in situ GC-MS (C_6-C_{10} hydrocarbons) and sampled in canisters and through DNPH cartridges (24h time resolution) to be analysed by GC-FID (C_2-C_6 hydrocarbons) and LC-UV (carbonyls), respectively. The total reactivity derived from the sum of individual OH reactivities for each measured compound was compared with total OH reactivity measured.

References:

4.078 Impacts of Emissions and Chemical Complexity on Global Simulations of Secondary Organic Aerosol.

Early Career Scientist

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Abstract:

Considering the importance of Secondary Organic Aerosol (SOA) to air quality and climate, the global budget remains uncertain with estimates of the global SOA source ranging several orders of magnitude. In addition to this, SOA schemes within global Chemical Transport Models (CTMs) significantly underestimate observed SOA. This is, in part, due to simplified SOA schemes and the absence of anthropogenic and biomass burning Volatile Organic Compound (VOC) emissions. We used the U.K Chemistry and Aerosol (UKCA) model to simulate SOA formation using the modal GLObal Model of Aerosol Processes (GLOMAP-mode) aerosol microphysics scheme. We assessed how emissions from various sources (biogenic, anthropogenic and biomass burning) and SOA schemes of varying chemical complexity (fixed-yield and 2-product) influence uncertainty in simulated SOA both regionally and globally. By comparing simulated SOA to Aerosol Mass Spectrometer (AMS) observations we were able to quantitatively evaluate model performance and constrain uncertainties in the simulated SOA budget. Initial results suggest uncertainty in the global SOA source is primarily due to which VOC species are included in the SOA scheme. We also find that combing the 2-product scheme with biogenic, anthropogenic and biomass burning VOC emissions the model skill at capturing the observed spatial pattern of SOA is enhanced. This suggests that both increased chemical complexity and more explicit representation of VOC chemistry within the SOA scheme are required for more accurate simulation of SOA. Significant differences in aerosol optical depth (AOD) and aerosol number concentration were also observed across these simulations, implying that uncertainties in SOA modelling has the potential to influence simulated climate. This work will be continued by considering uncertainties for future projections of SOA under climate change.
Carboxylic acid photochemistry is a marine source of glyoxal and other aldehydes.

Early Career Scientist

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Abstract:

Glyoxal is a highly water-soluble precursor in secondary organic aerosol (SOA) formation. Over land, glyoxal is known to be produced by the oxidation of volatile organic compounds (VOC) from both anthropogenic and natural sources. However, marine sources are still not well understood. Previous studies above the remote Pacific Ocean have detected typical glyoxal mixing ratios of 30-40 pptv, concentrations too high to be explained with current chemical understanding or atmospheric models. Because the lifetime of glyoxal is very short, the glyoxal found must have originated from, rather than been transported to, the open ocean. Furthermore, eddy covariance measurements indicate that an organic surface microlayer may be producing the glyoxal by as yet unknown processes. Here we present laboratory studies of the formation of glyoxal from carboxylic acid photochemistry. Nonanoic, octanoic, and heptanoic acids were floated on water in a quartz flow tube. Irradiation of the carboxylic acids converts them into their equivalent alkenals. The alkenals are volatile and are carried into a dark glass bottle by purified atmospheric gas. Subsequent ozonolysis of the alkenals in the glass bottle leads to the formation of glyoxal. We employ a PTR-MS to detect the alkenals, and a cavity-enhanced DOAS to detect the glyoxal. Considering the ubiquitous occurrence of carboxylic acids and their derivatives in the environment, this mechanism has the potential to be a significant source of glyoxal in the atmosphere.
Isoprene emissions account for about 50% of atmospheric non-methane reactive carbon. For this reason, assessing the role of biogenic emissions on secondary organic aerosol and ozone formation requires a complete understanding of the isoprene oxidation mechanism under a variety of nitrogen oxide (NO\textsubscript{x}) concentration regimes. The study of low-NO\textsubscript{x} isoprene oxidation products has recently been advanced by the synthesis of key isoprene oxidation products: isoprene hydroxyhydroperoxide (ISOPOOH) and isoprene epoxydiols (IEPOX). We present studies aimed at understanding the oxidation of these compounds in both the gas and aqueous phase. In the gas phase, the NO\textsubscript{x} dependence on the formation of small carbonyls including formaldehyde and glyoxal from the oxidation of ISOPOOH and IEPOX will be addressed. In the aqueous phase, studies will focus on the differences in the oxidation mechanism between gas and condensed phase,
the production of hydroxyl radicals (OH), OH reaction rates and particle formation. Together these experiments provide unique insights into the isoprene oxidation mechanism across both NO$_x$ regimes and phase boundaries.
4.082 Estimation of light absorption of brown carbon in PM2.5 with an improved AAE method.

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Abstract:

The strong spectral dependence of light absorption of brown carbon (BrC) aerosol is regarded to influence aerosol’s radiative forcing significantly. The Absorption Angstrom Exponent (AAE) method has been widely used in previous studies to attribute light absorption of BrC at shorter wavelengths for ambient aerosols, with a theoretical assumption that the AAE of “pure” black carbon (BC) aerosol equals to 1.0. In this study, the AAE method was applied to both urban and rural environments in the Pearl River Delta (PRD) region of China, with an improvement of constraining the realistic AAE of “pure” BC through statistical analysis of on-line measurement data. A three-wavelength photo-acoustic soot spectrometer (PASS-3) and aerosol mass spectrometers (AMS) were used to explore the relationship between the measured AAE and the relative abundance of organic aerosol to BC. The regression and extrapolation analysis revealed that more realistic AAE values for “pure” BC aerosol (AAE\textsubscript{BC}) were 0.86, 0.82, and 1.02 between 405 and 781 nm, and 0.70, 0.71, and 0.86 between 532 and 781 nm, in the campaigns of urban\textsubscript{winter}, urban\textsubscript{fall}, and rural\textsubscript{fall}, respectively. Roadway tunnel experiments were conducted and the results further confirmed the representativeness of the obtained AAE\textsubscript{BC} values for the urban environment. Finally, the average light absorption contributions of BrC (± relative uncertainties) at 405 nm were quantified to be 11.7%(±5%), 6.3%(±4%), and 12.1%(±7%) in the campaigns of urban\textsubscript{winter}, urban\textsubscript{fall}, and rural\textsubscript{fall}, respectively, and those at 532 nm were 10.0%(±2%), 4.1%(±3%), and 5.5%(±5%), respectively. The relatively higher BrC absorption contribution at 405 nm in the rural\textsubscript{fall} campaign could be reasonably attributed to the biomass burning events nearby, which was then directly supported by the biomass burning simulation experiments performed in this study.
4.083 Impacts of cloud aqueous processes on chemistry and transport of biogenic VOC.

Early Career Scientist

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Abstract:

Biogenic volatile organic compounds (BVOC) are crucial precursors in the formation of ozone and secondary organic aerosols, and serve as an important radical reservoir in the atmosphere. Recently, the role of aqueous processing of biogenic VOC has received attention, and convective transport and aqueous-phase processing in clouds can influence gas-phase reactivity and organic aerosol formation. We select a convective weather condition with non-precipitating cumulus clouds to understand the effects of cloud aqueous processes on vertical distributions of key BVOC species and their role on BVOC oxidation. We simulate the event utilizing the National Center for Atmospheric Research's Large Eddy Simulation (LES) model. Updated aqueous chemistry, including the oxidation of isoprene, methyl vinyl ketone, and methacrolein, and the formation and oxidation of acetic acid, glycolic acid, glyoxylic acid, pyruvic acid and oxalic acid, is coupled to the chemical mechanism in the LES model to examine the effect of the multi-phase chemistry. High-resolution LES modeling that resolves the strong turbulent environment and fast aqueous-phase reactions in clouds can accurately represent the cloud formation in the atmosphere and provide detailed turbulent transport in different regions of the cloud. Generally, the cloud formation induces upward transport of chemical species with downdrafts along the edges of clouds. We examine the role of physical turbulent processes with aqueous chemistry to investigate and explain the change in distributions and the OH reactivity of BVOC species in clouds. Besides turbulence-induced segregation for isoprene and OH near the surface, we find much stronger segregation and reaction rate reduction in clouds due to aqueous-phase chemistry, which can further affect ozone production and SOA formation time scales.
Revising concepts of methanesulfonic acid (MSA) formation in the remote tropical Pacific marine boundary layer using high-resolution measurements and a thermodynamic model of aerosol chemistry.

Early Career Scientist

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Abstract:

We report and interpret high-resolution methanesulfonic acid (MSA) and methanesulfonate (MS\textsuperscript{–}) measurements collected during a field campaign in the tropical Pacific marine boundary layer (MBL). An earlier study noted vapor-phase MSA was twice as concentrated near the ocean surface as at the top of the mixed layer. We use aerosol composition measurements and the online Extended AIM Aerosol Thermodynamics Model (E-AIM; Clegg et al. [1998]) to demonstrate that this pattern is due to evaporation of particulate MSA from sulfuric acid-dominated particles. We cannot rule out the possibility of a small homogeneous production source.

Up to 99% of the MS\textsuperscript{–} in aerosol forms in the aqueous phase after DMSO/MSIA vapor is wet-scavenged and oxidized to MS\textsuperscript{–} in aerosol water and cloud droplets. More alkaline coarse-mode aerosol generates MS\textsuperscript{–} by this mechanism and gathers MSA evading from fine particles. It is not clear which of these paths is more important. The lower FT is a source of MSA to the buffer layer (BuL) (entrainment flux: $9.9 \times 10^{11}$ molecules/cm\textsuperscript{2}/d), probably dissolving in cloud droplets and emerging below cloud as find-mode MS\textsuperscript{–}. Most fine-mode MS\textsuperscript{–}, however, is formed in cloud from DMSO/MSIA, while coarse-mode MS\textsuperscript{–} is formed on sea salt particles that have greater alkalinity.
On inferring the unobserved chemical state of the atmosphere: box model experiments.

Presenting Author:

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Abstract:

Chemical data assimilation in numerical models of the atmosphere has to deal with strongly non-linear interactions. Commonly assimilated observations (i.e. retrieved satellite products) exist for only a selected few of those key gas phase compounds (CO, O$_3$, NO$_2$, CH$_2$O), and assimilating those in models assuming linearity begs the question of: To what extent we can infer the remainder to create a new state of the atmosphere that is chemically sound and optimal? In our work we present systematic investigation of sensitivities that exist between chemical compounds under varying ambient conditions in order to inform scientists on the potential pitfalls when assimilating single/few chemical compounds into complex 3D chemistry transport models.

In order to do this, we developed a box-modeling assimilation tool (BOXMOX/BEATBOX) based on the Kinetic PreProcessor (KPP) in which we can conduct simulations with a suite of ‘mechanisms’, sets of differential equations describing atmospheric photochemistry. The box modeling approach allows us to sample a large variety of atmospheric conditions (urban, rural, biogenically dominated, biomass burning plumes) to capture the range of chemical conditions that typically exist in the atmosphere. Included in our suite are ‘lumped’ mechanisms typically used in regional and global chemistry transport models as well as the Master Chemical Mechanism (MCM). We will undertake an Observing System Simulation Experiment approach using the MCM prediction as ‘nature’ or ‘true’ state and assimilate idealized synthetic observations (from MCM) into different mechanism under various environments. Different chemical sensitivity approaches will be compared: 1) adjoint, 2) ensemble and 3) hybrid: using an ensemble of adjoint sensitivities, in order to derive gain matrices to optimally infer the unobserved part of the atmospheric chemical modeled states.
4.086 Reassessing the global secondary organic aerosol (SOA) budget and vertical distribution: stronger production, faster removal, shorter lifetime..

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Abstract:

Current chemistry climate models fail to reproduce the vertical distribution of organic aerosols (OA) with a large underprediction near the surface and a factor of 10-100 divergence in predicted OA profiles among models, reflecting our poor understanding of processes controlling the OA lifecycle. Recent laboratory and ambient measurements suggest that both production yields and removal rates of chemically produced secondary organic aerosols (SOA) are much stronger and more diverse than currently assumed in chemistry-climate models. In this study, we re-assess the global SOA distribution and budget with newly proposed SOA production and loss processes derived from these recent measurements, as well as from theoretical calculations. We evaluate and discuss the relative importance of increased production rates (wall corrected yields), chemistry of semi-volatile and intermediate volatility organic compounds, removal pathways for organic vapors and particles (e.g. dry and wet deposition, photo-dissociation, evaporation, and heterogeneous surface reactions), and their effect on the SOA vertical distribution and budget using a global chemistry-transport model. We compare simulated SOA from various model configurations against ground, aircraft and satellite measurements to assess the extent to which these new developments in our understanding of SOA formation and removal processes are consistent with observed characteristics of the SOA distribution. Our results show strong changes in predicted vertical profiles of organic aerosols with higher SOA concentrations in the boundary layer and lower concentrations in the upper troposphere, which appear to be in a better agreement with aircraft measurements.
4.087 Characterizing secondary organic aerosol yields from biogenic hydrocarbon precursor mixtures using an aerosol chemical ionization time-of-flight mass spectrometer.

Early Career Scientist

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Abstract:

We present a detailed characterization of an online method for simultaneous measurement of gas and aerosol chemical composition and abundance using chemical ionization high-resolution time-of-flight mass spectrometry equipped with an aerosol inlet (aerosol-CIMS). Characterization of the performance of the aerosol inlet shows unique volatilization profiles for a range of inorganic and organic aerosol with negligible thermophoretic effects under a centerline temperature of 330°C. To assess the performance of the aerosol inlet, a mixture of biogenic hydrocarbon precursors (α/β-pinene, limonene, sabinene) were oxidized via atmospheric gas-phase oxidation in an oxidative flow reactor and the gas and aerosol phase oxidation products were measured by aerosol-CIMS. As the temperature gradient of the aerosol inlet was increased, we observe changes in O/C ratios and highly functionalized species. We relate this observed, speciated chemistry to the volatility of the aerosol, and compare observed SOA yields to other methods. Comparison of aerosol mass measured by a single mobility scanning particle sizer and aerosol-CIMS suggests that while aerosol-CIMS is an important measurement technique for identifying aerosol chemical composition as it relates to aerosol physical properties like volatility, its ability to measure total organic aerosol is limited. Despite these limitations, multiple reagent ions (iodide, acetate) provide insight into total organic aerosol yields using aerosol-CIMS.
4.088 Uncertainties in Particle Wall Loss Correction during Secondary Organic Aerosol Formation in Chamber Experiments.

Early Career Scientist

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Abstract:

Particle wall deposition must be accounted for when determining secondary organic aerosol (SOA) yields during laboratory chamber experiments. However, a number of different particle wall loss correction methods exist, adding uncertainty to this correction. Furthermore, the use of high concentrations of seed particles has been suggested as a way to minimize the effects of vapor wall loss. Use of high seed particle concentrations complicates the particle wall loss correction, as coagulation will need to be accounted for. We present experiments and model simulations showing how particle wall loss rates change with different seed concentrations and size distributions and demonstrate how coagulation can be accounted for. We furthermore present alpha-pinene ozonolysis experiments using differing seed concentrations and size distributions. We apply four different particle wall loss correction methods to these experiments to quantify the uncertainty in this correction. This study demonstrates the importance of properly accounting for particle wall loss, especially when comparing SOA yields measured in different chambers using different particle wall loss correction methods.
Emissions from human activities have led to important changes in the composition of the atmosphere. Computational models are used to understand the impacts of emissions on air quality on local, regional, and global scales and the global climate system. An accurate representation of atmospheric chemistry is of critical importance in these models. The mission of the International Union of Pure & Applied Chemistry (IUPAC) Task Group on Atmospheric Chemical Kinetic Data Evaluation is to provide evaluated kinetic data for gas-phase, heterogeneous, and aqueous-phase reactions. More than 1000 datasheets are maintained on the website here http://iupac.pole-ether.fr. From the website, it is possible to join the mailing list, access datasheets, search the database (by species names, formula, InChI, or SMILES) and download summary tables. The Summary Tables provide kinetic data on:

- Gas-phase reactions of Ox, NOx, HOx, SOx, organic compounds, inorganic and organic halogens
- Heterogeneous reactions on solid surfaces
- Heterogeneous reactions on liquid surfaces.
The datasheets provide summaries of experimental data and procedures organized in four broad groups:

- Gas-phase reactions.
- Photolysis reactions.
- Heterogeneous reactions on solid substrates.
- Heterogeneous reactions on liquid substrates

Datasheets are being added for aqueous-phase reactions. Recommended data are published periodically in a special issue of Atmospheric Chemistry and Physics here http://www.atmos-chem-phys.net/special_issue8.html
5.001 Factors Affecting Aerosol Radiative Forcing.

Early Career Scientist

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Abstract:

Rapid industrial and economic growth has meant a large amount of aerosols in the atmosphere with strong radiative forcing (RF) upon the climate system. Aerosol RF is determined by emissions and various chemical-transport-radiative processes in the atmosphere, a multi-factor problem whose individual contributors have not been well quantified. In this study, we analyze the major factors affecting RF of secondary inorganic aerosols (SIOAs, including sulfate, nitrate and ammonium), primary organic aerosol (POA), and black carbon (BC). We analyze the RF of aerosols produced by 11 major regions across the globe, including but not limited to East Asia, Southeast Asia, South Asia, North America, and Western Europe. Factors analyzed include population size, per capita gross domestic production (GDP), emission intensity (i.e., emissions per unit GDP), chemical efficiency (i.e., mass per unit emissions) and radiative efficiency (i.e., RF per unit mass).

We find that among the 11 regions, East Asia produces the largest emissions and aerosol RF, due to relatively high emission intensity and a tremendous population size. Although Southeast Asia also has large emissions, its aerosol RF is alleviated by its lowest chemical efficiency. The chemical efficiency and radiative efficiency of BC produced by the Middle East-North Africa are the highest across the regions, whereas its RF is lowered by a small per capita GDP. Both North America and Western Europe have low emission intensity, compensating for the effects on RF of large population sizes and per capita GDP.

There has been a momentum to transfer industries to Southeast Asia and South Asia, and such transition is expected to continue in the coming years. The resulting relocation of emissions would mean drastic changes in both the spatial distribution and the magnitude of RF, with consequences on regional and global climate forcing.
Abstract:

Future change in surface ozone in a warming climate is an important question for the United States. Analyses of historical ozone change in response to climate change, although useful for validating theories regarding future ozone changes, are complicated by concurrent changes in anthropogenic emissions. Here we find that the individual contributions of climate and precursor emissions to US historical ozone changes over 1990–2014 can be distinguished by contrasting the changes in daytime versus nighttime ozone, based on an analysis of observed and simulated annual mean ozone time series. In particular, climate variability has determined ozone interannual variability, particularly for the daytime ozone, while reductions of anthropogenic NOx emissions have primarily driven an increase in the nighttime ozone. Our results have important implications for future ozone change studies and ozone mitigation.
5.005 Wildfires in a warmer climate: Emission fluxes, emission heights and black carbon concentrations in 2090-2099.

Early Career Scientist

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Abstract:

While there is consensus about a future decrease in carbonaceous aerosol emissions from anthropogenic sources, the future trend in aerosol emissions from wildfires is much more uncertain. Previous studies indicated that global warming could significantly increase wildfire activity in the 21st century, but the complex links between future changes in emission fluxes, emission heights and aerosol long-range transport remain to be quantified.

In this study, we use the process-based fire model SPITFIRE within the global vegetation model JSBACH to simulate wildfire activity for present day climate conditions and different future Representative Concentration Pathways (RCPs). Simulated fire emissions serve as input for the aerosol-climate model ECHAM6-HAM2, which has been extended by a semi-empirical plume height parametrization.

In our presentation, we will show that, compared to present day climate conditions, the modelled changes in emission fluxes for the period 2090-2099 are most pronounced for the strongest warming scenario RCP8.5 (-37% in the tropics, +49% in the extra-tropics). Tropospheric Black Carbon (BC) concentrations are similarly affected by changes in emission fluxes and changes in climate conditions with regional relative variations of -50% to +100%. In the vicinity of the major extra-tropical biomass burning regions the enhanced wildfire emission fluxes in turn introduce a distinct increase in Aerosol Optical Thickness (AOT). Due to the compensating effects of fire intensification and more stable atmospheric conditions, simulated future changes in mean emission heights do not exceed 0.3km.

Overall, we conclude that the radiative impact of the expected future increase in wildfire activity is of a similar magnitude like the opposed radiative impact introduced by a decrease in anthropogenic BC emissions. Changes in emission heights, however, are of minor importance.

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Abstract:

The spatial distribution of aerosols and their chemical composition dictates whether aerosols have a cooling or a warming effect on the climate system. Hence, properly modeling the 3-dimensional distribution of aerosols is a crucial step for coherent climate simulations. Since surface networks only give 2-D data, and most satellites supply integrated column information, it is thus important to integrate aircraft measurements in climate model evaluations. In this study, the vertical distribution of secondary inorganic aerosol (i.e. sulfate, ammonium and nitrate) is evaluated against a collection of 14 AMS flight campaigns and surface measurements from 2000-2010 in the USA and Europe.

GISS ModelE2 is used with multiple aerosol microphysics (MATRIX, OMA) and thermodynamic (ISORROPIA II, EQSAM) configurations. Our results show that the MATRIX microphysical scheme improves the model performance for sulfate, but that there is a systematic underestimation of ammonium and nitrate over the USA and Europe in all model configurations. In terms of gaseous precursors, nitric acid concentrations are largely underestimated at the surface while overestimated in the higher levels of the model, influenced by strong stratosphere-troposphere exchange. Heterogeneous reactions on dust surfaces is an important sink for nitric acid, even high in the troposphere. At high altitudes, nitrate formation is calculated to be ammonia limited. The underestimation of ammonium and nitrate in polluted regions is most likely caused by a too simplified treatment of the NH3/NH4+ partitioning which affects the HNO3/NO3-partitioning.
5.008 Aerosols and climate interaction in Community Earth System Model.

Early Career Scientist

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Abstract:

Two environmental challenges that the world faces today are air pollutions (such as particulate matters or aerosols) and climate change. Using NCAR’s global climate model (CESM1) that couples an online aerosol module, we studied how these two problems are entangled.

Firstly, on aerosol effects on climate, we showed that the decline of aerosol pollutions projected in the next few decades will lead to additional warming in the Northern Hemisphere, and longer and stronger heat extremes in the tropics. Surprisingly, per degree of global warming, the role of aerosols in causing warming even outweighs greenhouse gases over some regions, due to stronger changes in atmospheric circulations.

Secondly, on climate effects on aerosols, we continue to show that global warming in general leads to heavier pollution concentration in the atmosphere, even at current emission level. This suggests that air pollution regulations needs to be further strengthened to meet the current air quality standard. The results are counterintuitive because the total rainfall that removes aerosols from the air is increasing under global warming conditions. The underlying mechanisms for such a warming-driven pollution rise were investigated using CESM1.
5.011 Changes in Air Quality in Different World Regions for the Past Decades: analyses using chemistry-climate simulations and observations from satellite and monitoring stations.

Early Career Scientist

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Abstract:

Surface emissions of atmospheric compounds have changed dramatically in many world regions during the past decades. In this study, we investigate the spatial variability of long-term changes of atmospheric compounds such as carbon monoxide (CO) and nitrogen dioxide (NO$_2$) over the 1980-2010 period, as simulated by the Community Atmospheric Model (CAM4-Chem). Simulated trends are compared with temporal changes derived from different satellite and ground-based observations, with a focus on Europe, North America and Eastern China regions. Results of simulations using the Hadley Centre Coupled Model-United Kingdom Chemistry and Aerosols model (HadGEM3-UKCA) are included in the analyses.

Similar negative trends in the CO and NO$_2$ tropospheric columns are generally observed in both CAM4-Chem and HadGEM3-UKCA simulations and measurements, especially in Europe, North America and Eastern China regions. During the 2000-2010 period, CAM4-Chem simulated an increase in CO column trends while MOPITT reported a decrease in Eastern China, in contrast with the significant increase in anthropogenic CO emissions during this period. Such
differences could be linked to several factors such as uncertainties in the chemical dynamical schemes included in the model, as well as inaccuracies in the satellite retrievals, or in the surface emissions, which mainly drive the CO trends. Temporal changes in tropospheric NO$_2$ columns from model simulations were consistent with satellite observations as well as surface measurements from the US-EPA and EMEP networks in Europe and the USA. However, the magnitudes of modeled trends were generally lower than those from satellite and surface observations.
Abstract:

Black carbon (BC) has been identified as the second most important anthropogenic global warming agent by virtue of its strong absorption of solar radiation and significant reduction of snow albedo after its deposition. Observations have shown substantial and accelerating snow melting over high mountains and the Arctic associated with BC particles deposited on snow (“dirty snow”). Previous studies have investigated the effects of such factors as BC particle-snow grain mixing state, BC particle size, and snow grain size on BC-induced snow albedo feedback that accelerates snowmelt with global climate change implications. However, two critical features involving 1) BC-snow multiple internal mixing and 2) snow grain shapes have not been accounted for in evaluating the BC-snow interactions and feedback. In this study, we develop a physically-based parameterization that accounts for the two aforementioned features based on a stochastic snow model to quantify BC-snow albedo feedback. Our preliminary results show that BC-snow multiple internal mixing enhances snow albedo reduction by 40-60% compared with external mixing, while spherical snow grains enhances the albedo reduction by 20-40% relative to Koch snowflakes and hexagonal-plate snow grains. The parameterization will be further incorporated into a widely used land surface model (the Noah-MP model), where the aerosol-snow-radiation interactions and feedback are currently absent, to improve snow albedo simulations associated with weather and climate models on multiple scales. Similar parameterization will also be done to account for dust-snow albedo feedback in our future studies.
5.013 Changes in anthropogenic surface emissions in different world regions during the past decades.

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Abstract:

Information on surface emissions on an accurate, timely and accessible basis is critical for understanding the current composition of the atmosphere, as well as for hindcast and forecast simulations. Different research teams worked over the past years to better quantify surface emissions, and to develop anthropogenic spatial and temporal distributions for different periods and regions. These inventories provide either emissions on a national basis for different countries, or gridded emissions at the global or regional scale.

We will present an evaluation of the most recent emissions datasets providing emissions for the 1960-2013 period, for different gaseous and particulate compounds, i.e. carbon monoxide, nitrogen oxides, volatile organic compounds, sulfur dioxide, ammonia, black and organic carbon, and particulate matter (PM10 and PM2.5).

The quality of the different emission datasets is difficult to assess. The methodology, input data, assumptions and proxies vary strongly in space and time among the inventories. We will discuss the consistency between global and regional inventories, as well as between the different chemical compounds, for 22 world regions. This work will help quantifying the uncertainties on anthropogenic emissions in the different regions.

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Abstract:

Accurate, evaluated, timely, and accessible emissions information is critical for understanding and making predictions about the atmosphere. We present recent progress of the Global Emissions Initiative (GEIA, http://www.geiacenter.org/), a community-driven joint activity of IGAC, iLEAPS, and AIMES. For the past quarter century, GEIA has served as a forum for the exchange of expertise and information on anthropogenic and natural emissions of trace gases and aerosols. GEIA supports a worldwide network of emissions data developers and users, providing a solid scientific foundation for atmospheric chemistry research. GEIA serves as a bridge between the environmental science, regulatory, assessment, policy, and operational communities. GEIA’s core activities include 1) facilitating analysis that improves the scientific basis for emissions data, 2) enhancing access to emissions information, and 3) strengthening linkages within the international emissions community. We highlight GEIA’s current work distributing emissions data, organizing the development of new emissions datasets, facilitating regional emissions studies, and initiating analyses aimed at improving emissions information. GEIA welcomes new partnerships that advance emissions knowledge for the future.
5.017 BC enhancement of glacier melting on Cerro Tronador: modeling and measurements.

Early Career Scientist

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Abstract:

The influence of Black Carbon (BC) on glacier melting is of great interest currently (1), but there are ongoing discussions on its magnitude, due to controversy on the appropriate methodology of simulation of this effect. Specially, there is a lack of sufficient experimental data regarding different physical and chemical processes involved in the aforementioned interaction (1, 2). Hence, online modules that incorporate this effect in general atmospheric circulation models still provide diverging results.

I will show preliminary results of the implementation of an online coupling between BC deposition and glacier enhanced melting, into the atmospheric transport model CCATT-BRAMS (3). We have already validated a meteorological simulation of CCATT-BRAMS in the Argentinean Andes (4). I will describe the most relevant direct and indirect effects of BC deposition on ice and snow, and the weaknesses of the available models that we wish to improve. I will also show preliminary results of the field campaign in glaciers located in Cerro Tronador, Argentina, including measurements of surface albedo and BC, mineral dust and volcanic ashes concentration (surface and vertical profile). We will assess the differences on the BC concentrations determined with different techniques (reflectometry, thermal-optical methods and incandescence methods) considering the interference of other particles. We will use the experimental data to get an alternative estimate of the BC enhanced melting. Additionally, we will try to use these results to validate or adjust BC emissions, deposition, post-depositional processes, albedo and enhanced melting in CCATT-BRAMS.

References
**5.018 Sensitivity of dust emissions to aerosol feedback and the impact of dust loading on climate forcing with varied resolutions using FIM-Chem.**

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Abstract:

Meteorological conditions directly impact aerosol loading, especially dust emissions. Variations in dust emissions on the other hand, will also impact meteorology and climate through direct and indirect aerosol forcing. To study these impacts in more detail we use the global Flow-following finite-volume Icosahedra Model (FIM, http://fim.noaa.gov/), a new global weather prediction model currently under development in the Global Systems Division of NOAA/ESRL, as it is coupled online with the aerosol modules from the Goddard Gobal Ozone Chemistry Aerosol Radiation and Transport (GOCART) model (FIM-Chem). FIM-Chem includes direct and semi direct feedback, and uses the dust schemes of GOCART and the Air Force Weather Agency (AFWA). FIM-Chem is able to investigate the contribution of climate feedbacks to simulated hyperspectral data by considering a range of simulations with different dust emissions and different levels of aerosol feedbacks enabled at four different spatial resolutions. The emitted dust flux and total emissions are highly depending on the wind, soil moisture and model resolution. We compare the dust emissions by including and excluding the aerosol radiative feedback in the simulations to quantify the sensitivity of dust emissions to aerosol feedback. The results show that all aerosol-induced dust emissions increase about 10% globally, which is mainly dominated by the contributions of anthropogenic black carbon (EC) aerosol. While the dust-induced percentage changes of dust emissions are about -5.5%, that indicates reduction effect globally. Also, the simulations based on different resolutions of 240x240 km, 120x120 km, 60x60 km and 30x30 km are performed to test the impacts of model resolution on total dust emissions. By comparing the dust emission sensitivity to aerosol feedback and model resolution, we can estimate the uncertainty of model resolution versus aerosol feedback. We also conduct FIM-Chem simulations to investigate the sensitivity of dust climate forcing to different model resolutions.
5.020 Diagnosing changes in free tropospheric ozone over Europe: A model study of past and future changes.

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Abstract:

In recent decades, the negative impacts of tropospheric ozone on human and ecosystem health have led to policy changes aimed at reducing emissions of ozone precursor gases such as nitrogen oxides (NO\textsubscript{x}) and volatile organic compounds (VOCs). Although emissions of these species have significantly decreased in Europe and North America since the early 1990s, observational data indicate that free tropospheric ozone over Europe has not decreased as expected. Uncertainty remains as to how much of a role the transport of stratospheric ozone or tropospheric ozone from remote source regions has played in recent trends, as well as how this will evolve in a changing climate.

The global chemistry-climate model SOCOL (SOlar Climate Ozone Links) is used to investigate tropospheric ozone over Europe from 1960 to 2100. To fully disentangle the effects of both long-range transport and input from the stratosphere, simulations are run with ozone tracers from 29 different atmospheric regions. In addition to a standard reference run, two sensitivity simulations are run: one with emissions of NO\textsubscript{x} and VOCs held constant at 1960 levels, one with methane (CH\textsubscript{4}) held at constant 1960 levels (in addition to the NO\textsubscript{x} and VOCs). Results suggest that the largest contributions to European tropospheric ozone originate from the tropical and northern mid-latitude boundary layer and free troposphere. Contributions from these regions increase over the historical period (1960-2010), indicating that changes in source gas emissions have affected ozone concentrations in the European free troposphere most strongly. Contributions from these regions then decrease from 2010-2100, but remain considerably larger than input from the stratosphere, which is relatively small in all simulations. The stratospheric contribution does, however, increase over the 21st century, in tandem with ozone recovery and a simulated strengthening of the Brewer Dobson circulation.
Aerosol and trace gases in the upper troposphere and lower stratosphere (UTLS) are important to the climate but still remain relatively less known to the science community. Recently NOAA-ESRL conducted a few filed campaigns to study in-situ aerosol and size distribution in the UTLS of Asia and North America. A sectional aerosol model coupled with community earth system model is used to investigate stratospheric composition, size distributions, optical properties of aerosols. Our model can reproduce aerosols composition and size distribution in the global UTLS compared with multiple field campaigns and other datasets (e.g. Mauna Loa Lidar, University of Wyoming aerosol size distributions). We found organics makes up to 40% of global stratospheric aerosol budget. Based on our model, we provides estimation of radiative forcing coming from stratospheric non-volcanic aerosols (including both sulfate and organics) since 1850.
5.023 Impacts of atmospheric and aerosol processes on black carbon concentrations in the Arctic.

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Abstract:
This study quantifies black carbon (BC) processes in three global climate models and one chemistry transport model, with focus on the seasonality of BC transport, emissions, wet and dry deposition in the Arctic. In the models, transport of BC to the Arctic from lower latitudes is the major BC source for this region. Arctic emissions are very small. All models simulated a similar annual cycle of BC transport from lower latitudes to the Arctic, with maximum transport occurring in July. Substantial differences were found in simulated BC burdens and vertical distributions, with CanAM (NorESM) producing the strongest (weakest) seasonal cycle. CanAM also has the shortest annual mean residence time for BC in the Arctic followed by SMHI-MATCH, CESM and NorESM. Overall, considerable differences in wet deposition efficiencies in the models exist and are a leading cause of differences in simulated BC burdens. Results from model sensitivity experiments indicate that convective scavenging outside Arctic reduces the mean altitude of BC residing in the Arctic, making it more susceptible to scavenging by stratiform (layer) clouds in the Arctic. Consequently, scavenging of BC in convective clouds outside the Arctic acts to substantially increase the overall efficiency of BC wet deposition in the Arctic, which leads to low BC burdens and a more pronounced seasonal cycle compared to simulations without convective BC scavenging. In contrast, the simulated seasonality of BC concentrations in the upper troposphere is only weakly influenced by wet deposition in stratiform clouds whereas lower tropospheric concentrations are highly sensitive.
5.025 Absorption coefficient of urban aerosol in Nanjing, west Yangtze River Delta of China.

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Abstract:

Aerosol absorption coefficient (AAC) is an indicator to assess the impact of absorbing aerosols on radiative forcing. In this study, the near-surface AAC and absorption angstrom exponent (AAE) in urban Nanjing, China, are characterized on the basis of measurements in 2012 and 2013 using the 7-channel Aethalometer (model AE-31, Magee Scientific, USA). The AAC is estimated with direct and indirect corrections, which show consistent temporal variations and magnitudes at 532 nm. The mean AAC at 532 nm is about 43.23±28.13 Mm$^{-1}$ in urban Nanjing, which is much lower than that in Pearl River Delta and as the same as that in rural areas (Lin'an) in Yangtze River Delta. The AAC in urban Nanjing shows strong seasonality (diurnal variations), high in cold seasons (at rush hours) and low in summer (in afternoon). It also show synoptic and quasi-two-week cycles in response to weather systems. Its frequency distribution follows a typical lognormal pattern. The 532 nm-AAC ranging from 15 to 65 Mm$^{-1}$ dominates, accounting for more than 72% of the total data samples in the entire study period. Frequent high pollution episodes, such as those observed in June 2012 and in winter 2013, greatly enhanced AAC and altered its temporal variations and frequency distributions. These episodes are mostly due to local emissions and regional pollutions. Air masses from northern China to Nanjing can sometimes be highly polluted and lead to high AAC at the site. AAE at 660/470 nm from the Schmid correction (Schmid et al., 2006) is about 1.56. Low AAEs mainly appear in summer in response to the relative humidity (RH). AAC increases with increasing AAE at a fixed aerosol loading. The RH-AAC relationship is more complex. Overall, AAC peaks around RH values of 40% (1.31.6), and 80% (1.3
5.027 Impact of aerosols on regional climate in southern and northern China during strong/weak East Asian summer monsoon years.

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Abstract:

In this work, we mainly simulate the effects of aerosols on regional climate in southern (SC) and northern China (NC), and compares the differences of aerosol climatic effects in strong/weak summer monsoon years with a modified regional climate model RegCM4. The results show that the total climatic effects of aerosols cause the decline of averaged air temperature and precipitation of SC and NC in summer. In NC, the strength of temperature drop in strong summer monsoon years is higher than that in weak summer monsoon years, indicating the possible impact from the different changes of radiation, circulation, and precipitation. The decrease of precipitation is more significant in NC in weak summer monsoon years, while it is stronger in SC in strong summer monsoon years due to the difference of aerosol distribution as well as the effects on circulation and cloud microphysics processes. Besides, aerosol effects also cause a decrease of zonal wind at 850hPa in SC and an increase in NC. The cooling center is more northerly and stronger in strong summer monsoon years, which results in the differences of vertical circulation anomaly and meridional wind anomaly at 850hPa. In weak summer monsoon years, meridional wind at 850hPa is increased in NC, while it is found to be decreased in SC. In strong summer monsoon years, meridional winds at 850hPa in both NC and SC are weakened. However, the decrease in SC is much more distinctly and clearly.
Climate forcing of different secondary organic aerosol descriptions in the Community Earth System Model (CESM).

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Abstract:

We use the Community Earth System Model (CESM) to investigate the importance of different descriptions of particulate organic matter (OM) for climate simulations. We compare different schemes for the production of secondary organic aerosols (SOA). The simple scheme that only emits SOA gas (SOAG) based on assumptions on yields of Volatile Organic Compounds (VOC) is compared to a scheme that includes the Volatility Basis Set (VBS) to represent the aging process of VOCs during the path of formation of SOAs. This scheme parameterizes intermediates, following Hodzic et al. (2015), which includes newly available constraints from laboratory measurements (wall-corrected yields) and explicit modeling on both production and removal of organic aerosols. We also derive a medium complex VBS approach that is cheaper for climate model simulations. For the different model version, we will identify the climate forcing (direct and indirect), i.e. through aerosol-radiation interaction and aerosol-cloud interaction throughout the 20th century. The different descriptions will be evaluated with observations from recent aircraft campaigns.

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Abstract:

We present chemistry-climate modelling simulation results to investigate tropospheric and lower-stratospheric ozone trends and variability from 1987 to 2014 observed at Lauder, New Zealand, which is a Southern Hemisphere (SH) background site. We examine impacts of changes in meteorology, ozone precursors, ozone depleting substances, and greenhouse gases on ozone, based on several model simulations including those conducted for the Chemistry Climate Modelling Initiative (CCMI). In the troposphere, ozone varies mainly with meteorology and ozone precursors, whereas dynamical changes contribute mainly to lower-stratospheric ozone changes.
5.031 Drivers of Increase in Atmospheric Methane since 2007.

Early Career Scientist

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Abstract:

Atmospheric methane (CH$_4$) is now the second most important anthropogenic greenhouse gas in terms of radiative forcing (IPCC, 2013). After continuously increasing, methane levels plateaued from 1999 to 2006, but since then have continued to increase (Sussmann et al., 2012). At the same time, $\delta^{13}$C have decreased globally (Nisbet et al., 2014). The contribution of increasing natural and anthropogenic emissions from agriculture and fossil fuel remain unknown.

Simulations forced with prescribed meteorological fields have been performed for the 2001-2010 period using the global chemistry-climate model (CCM) SOCOLv3 (Stenke et al., 2013). 48 methane tracers have been included in the model and used together with flux boundary conditions for CH$_4$ to allow the tracking of methane emissions from different source categories, (such as wetlands, rice paddies, ruminants, industry, etc.) as well as geographical regions. Isotopes of methane $^{13}$CH$_4$ have been implemented to simulate the trend in $\delta^{13}$C for the studied 2001-2010 time period.

An analysis of the implemented tracers elucidates the impact of different emission source categories by comparing the change between 2008-2010 and 2001-2003. 8 locations have been chosen from the NOAA ESRL global network for comparison. For all locations, a study of the change in contribution of the tracers as well as the $\delta^{13}$C trend indicates that a combination of increasing emissions over Africa, China, India, the Middle East, South America, and tropical Asia explain the observed rise in methane and can not be compensated by a negative contribution of decreasing emissions from Eurasia and Northern America and can explain the increase after 2007. Emissions over Africa, India and South America are mainly driven by agriculture, this, combined with decreasing fossil fuel emissions from Eurasia and North America, it explains the decrease in $\delta^{13}$C both observed and simulated.
5.034 Understanding and Quantifying the Missing Free Tropospheric Aerosol Loading Over Monsoon Asia.

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Abstract:  
Black Carbon (BC) is a short-lived, heterogeneously distributed, species impacting radiative forcing and the atmospheric energy balance. Over Southeast Asia, its dominant sources are rapid and growing urbanization and massive forest and agricultural fires. A recent pair of papers used a time-varying Kalman Filter method and multiple ground and remotely sensed measurements to show both sources contribute to the underestimation. However, the weekly and monthly scale changes in the spatial, temporal, and vertical distribution are still not well quantified. In addition, there is considerable in-situ dynamical and chemical processing contributing to the underestimation. It is known that the BC in this region is heavily chemically aged/mixed and heterogeneously distributed in space and time, due to urban-scale chemistry, heat from fires, intense convection, and high atmospheric water loadings.  
This work uses a combination of chemical and optical measurements from both surface and remotely sensed platforms, in combination with fundamental physical and atmospheric chemical models, to quantify the timing and amount of emissions, aging, and method of the transport of the aerosols. Measurements are taken from multiple platforms, and look at multiple species, directly and indirectly related to BC, including: fires, extinction, AOD, and trace gasses.  
Comparisons of the inverse-modeled results are shown to match well under both El Nino (2006) and typical (2010) years. Furthermore, the signal can be observed by more precise instruments far away from the source regions.  
A sensitivity analysis is performed and used to quantify the drivers behind the underestimate in the free tropospheric loading. It is found that a combination of underestimated fires, convective feedback, underestimated emissions, and insufficient aging are all responsible. Applications of these results to other Monsoon regions will be discussed.
5.035 The Underestimated Role of the Stratospheric Source on Tropospheric Ozone.

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Abstract:

The atmospheric composition is strongly influenced by changing atmospheric dynamics, in potential relation to climate change. A prominent example is doubling of the stratospheric influence on ozone at the summit station Zugspitze (2962 m a.s.l., Garmisch-Partenkirchen, Germany) between the mid-seventies and 2005, roughly from 11 ppb to 20 ppb. Systematic efforts for identifying and quantifying this influence have been made since the late 1990s (e.g., H. E. Scheel in: http://www.forst.tu-muenchen.de/EXT/LST/METEO/staccato (2002)) based on data filtering ozone, relative-humidity (RH) and 7Be measurements. Meanwhile, routine lidar measurements of ozone and water vapour carried out since 2007, combined with radiosonde data and trajectory calculations, have revealed the presence of stratospheric intrusion layers on up to 80 % of the yearly measurement days. The pronounced seasonal cycle for deep intrusions disappears if one looks at the entire free troposphere, which confirms the results of Beekmann et al. (J. Atmos. Chem. 28 (1997), 29-44). The mid- and upper-tropospheric intrusion layers seem to be dominated by very long downward transport up to a full tour around the northern hemisphere. These layers remain considerably dry, typically with RH ≤ 5 % at the centre of the intrusion. In 59 % of the deep intrusion cases with subsidence times up to six days the minimum relative humidity (RH) was 1 % or less (Trickl et al., Atmos. Chem. Phys. 14 (2014), 9941-9961). It is interesting to note that, in recent years, most pronounced ozone maxima have been related to a stratospheric origin rather than to long-range transport from remote boundary layers. It is difficult to answer the question if this fact is related to improving air quality in the most relevant source regions.
What does a 1.5 °C warmer world mean for atmospheric chemistry?

Early Career Scientist

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Abstract:

One aim to come from the recent COP21 negotiations in Paris was to keep global mean
temperature rise this century to well below 2 °C above pre-industrial levels, and to drive
efforts towards limiting the temperature increase to 1.5 °C. Our work considers the
potential impact of such targets on atmospheric chemistry, including changes in natural
emissions from vegetation and lightning, deposition fluxes, stratospheric and
meteorological changes that impact air quality (the “climate penalty”). To do this, we
draw on the large archive of climate model data from the Coupled Model Intercomparison
Project, phase 5 (CMIP5) to examine the spread and robustness of the results. In addition,
we demonstrate enhanced climate model agreement when comparing temperature
targets (e.g. 2 °C) rather than dates (e.g. 2070-2100).
5.037 Temperature as a driver of ground-level ozone concentration in Europe.

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Abstract:
The link between high temperature and extreme concentrations of ozone is generally well established. In this work, we employ a suite of statistical modelling techniques to systematically examine the importance of temperature along with a number of other local meteorological parameters such as humidity, solar radiation intensity, as well as wind speed and direction for predicting observed ozone concentrations in Europe. Moreover, the influence of large-scale circulation is also investigated through a set of airflow indices. We show that temperature is a significant driver of summertime ozone concentration over much of the western-to-central European mainland, however this relationship does not apply for much of the rest of Europe, where a simple persistence relationship has more skill, and parameters other than temperature are often ranked higher as predictors. The extreme components of the ozone concentration frequency distribution show similar patterns to the full distribution. We employ the same suite of statistical modelling techniques as employed in our analysis of the observations to examine the driving parameters in regional chemistry-climate model simulations. Using our regional chemistry-climate model and a detailed chemical box model, we investigate this ozone-temperature relationship using several commonly-used chemical mechanisms. We show that the temperature dependence of the ozone production rate can be broken down into a component due to the temperature dependence of isoprene emissions, and a component due to temperature dependent reaction rates. Geographically, the temperature dependence of ozone production rate is related to the chemical regime (NOx vs. VOC sensitive chemistry).
5.038 The application of long-term observations of NOx and CO to constrain a global emissions inventory.

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Abstract:

Accurate knowledge of tropospheric ozone is important for understanding its current and future effects on human health, air quality, and climate. However, global chemistry-climate models generally have problems reproducing tropospheric ozone concentrations, seasonal cycles and interannual trends. Successful tropospheric ozone simulations require high quality information on the emissions of ozone precursors, including nitrogen oxides (NOx), carbon monoxide (CO) and volatile organic compounds (VOCs). We analyzed CO and NOx measurements from four megacities (Los Angeles, New York City, London, and Paris). It has been shown that CO and VOC emissions are highly correlated in urban areas. However, CO is more extensively measured than VOCs. Therefore, we compared the long-term evolution of the measured NOx/CO enhancement ratio in each city to the ratio of the emissions of these two pollutants reported by the MACCity global emissions inventory at the inventory grid points nearest the city. The longest available measurement record (~50 years) is from Los Angeles, where the measured NOx/CO ratios are consistently smaller than the emission ratio in the MACCity inventory and the slope of the long-term trend in measured NOx/CO ratios is of significantly larger magnitude than for MACCity. The other three cities do not have as long of a data record, but the evolution of their NOx/CO ratios also implies that the
MACCity NOx/CO emissions trends are not steep enough. However, the agreement between the measured and inventory ratios is better for the shorter time period where measurements are available in these three cities. Comparisons of MACCity with fuel-based emission estimates for Los Angeles (for the 1990s and 2000s) suggest that the CO emissions are underestimated in the inventory, which results in the higher inventory NOx/CO ratios.
5.039 Drivers of global chemical climate forcing 1990 - 2010.

Early Career Scientist

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Abstract:

In the last few decades, Earth’s atmosphere has experienced large-scale changes in the concentrations and distributions of the short-lived climate pollutants (SLCPs) ozone, methane, and aerosols, driving an imbalance in Earth’s radiation budget. The recent changes in atmospheric chemical composition are driven by a number of inter-related processes, including changes in: the distribution and magnitude of emissions of reactive precursor pollutants; global land cover; atmospheric CO$_2$ concentration; and physical climate. We apply multiple observational datasets and a global carbon-chemistry-climate model (NASA ModelE2-YIBs) to quantify the magnitude of recent SLCP concentration changes and the associated radiative forcing for each chemical species over the period 1990 – 2010. Attribution studies evaluate the contributions of the various drivers (precursor emissions, land cover, and climate) to chemical forcing. We additionally quantify the forcing induced by SLCP changes resulting from changes in dry deposition and biogenic volatile organic compound emissions driven by exposure of the biosphere to enhanced atmospheric CO$_2$ concentrations over this period. NASA ModelE2-YIBs features fully interactive gas and aerosol chemistry, an interactive land carbon cycle, and a BVOC emission algorithm in which isoprene production is biochemically linked to photosynthesis. Time-slice simulations are forced with monthly anthropogenic and biomass burning air pollution emissions from the MACCity inventory and nudged with large-scale winds from the NASA GMAO MERRA reanalysis dataset. The calculated global-average radiative forcing > 0.1 W/m$^2$ from changes in the SLCPs over the period 1990 – 2010 is approximately 25% of the magnitude of the forcing from CO$_2$ concentration change over this period and is primarily driven by changes in reactive anthropogenic precursor emissions. In the mid- to upper-troposphere, though, climate change has a larger influence on ozone concentrations than do emission changes. The simulated climate state and atmospheric chemical composition are evaluated using reanalysis data, MODIS AOD, and TES ozone concentrations.
5.040 Contribution of Arctic seabird-colony ammonia to atmospheric particles and cloud-albedo radiative effect.

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Abstract:

The Arctic region is vulnerable to climate change and has been warming more rapidly than the global-mean rate for the Earth. Atmospheric particles and clouds play key, but complex roles in offsetting the radiative forcing from greenhouse gases. In the summertime, the Arctic atmosphere is pristine and strongly influenced by natural regional emissions, which have poorly understood climate impacts related to atmospheric particles and clouds. Recent evidence indicates that ammonia emissions from migratory-seabird guano are a primary contributor to summertime boundary-layer ammonia levels measured in the Canadian Arctic Archipelago region. In this study, we examine the impact of pan-Arctic seabird-colony ammonia on particle number concentrations, and the cloud-albedo radiative effect. Ammonia from seabird-colony guano is found to be a key factor contributing to bursts of newly formed particles, which are observed every summer in the near-surface atmosphere at Alert, Nunavut, Canada. Our GEOS-Chem-TOMAS chemical-transport model simulations indicate that the pan-Arctic seabird-influenced particles can grow by sulphuric-acid and organic vapour condensation to diameters sufficiently large to promote pan-Arctic cloud droplet formation in the clean Arctic summertime. We calculate that the resultant cooling tendencies could be large, exceeding \(-1 \text{ W m}^{-2}\) near the largest seabird colonies due to the effects of seabird-
influenced particles on cloud albedo. These coupled ecological-chemical processes may be susceptible to Arctic warming and industrialization.
CMIP5 estimate of Earth’s energy budget.

Early Career Scientist

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Abstract:
The inability to make direct measurements and the complexity of the processes involved make it difficult to quantify all the components of the Earth’s energy budget. Climate models have the potential to greatly inform our understanding of the relevant processes controlling the energy budget. There have been a series of climate model intercomparison projects (CMIP), the latest version of these projects, CMIP5, has contributions from dozens of global climate models. We use these models to understand how much energy has come into the Earth system since preindustrial times and the fate of that energy, whether it has warmed the oceans or been radiated back to space. Climate models have errors, biases and drifts that must be accurately accounted for to estimate anthropogenic climate change. Much of the drift can be accounted for by using anomalies from preindustrial control simulations. Despite these adjustments, there is substantial variation between the various CMIP5 models of fundamental quantities such as ocean heat content and top of the atmosphere radiative imbalance. However, much of this variation is around a mean that is consistent with observations. One quantity of interest is the ratio of energy radiated back to space to that absorbed by the ocean. Since 1957, the CMIP5 models estimate that roughly 63% of the integrated forcing is radiated back to space with 37% being stored in the ocean. The models further estimate that 15% of the change in the ocean heat content is stored in the deep ocean below 2000 m. With proper adjustments and corrections, the CMIP5 suite of climate models can robustly predict the historical trends and magnitudes of climate change. The success of these models to predict historical changes lends credence to their predictions of an ever changing future Earth.
5.042 Quantifying the frequency and duration of U.S. regional pollution episodes with EOF analysis: Model evaluation and projected 21st century changes.

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Abstract:

Effective planning and implementation of efforts to achieve the U.S. National Ambient Air Quality Standards for ozone and particulate matter should be underpinned by quantitative knowledge of pollutant responses to changes in climate and emissions. In the coming decade, computational advances will increasingly permit large ensembles to be generated from chemistry-climate and air quality models. These simulations span a range of possible emission and climate states, requiring statistical approaches for data reduction and for evaluating model skill at reproducing observed pollutant distributions and their variability that do not rely on exact space-time matching. Here, we propose an EOF approach to address these needs, and demonstrate a proof-of-concept application for a rapid screening tool to identify high pollution events and changes in their frequency and duration over time in ensemble simulations. We conduct an EOF analysis on a $1^\circ \times 1^\circ$ gridded product for maximum daily 8-hour (MDA8) ground-level ozone from 1993-2013, interpolated from the U.S. EPA CASTNet, AQS, and Canadian CAPMon networks that was previously applied to evaluate the spatial extent of pollution episodes in chemistry-climate models. We focus on summertime and retain the first 5 EOFs that explain 73% of the total variance, and subject them to Varimax rotation. We apply the same analysis to surface MDA8 ozone archived from three sets of simulations with the GFDL chemistry-climate model: (1) GFDL AM3 nudged to NCEP re-analysis for 1993-2007, (2) GFDL AM3 free-running, forced with observed sea surface temperatures and sea ice (3) GFDL CM3 (AM3 with a fully coupled ocean) 21st century simulations. We find EOFs (spatial patterns) in all simulations are consistent with those derived from observations. We will demonstrate statistical approaches to extract information regarding temporal changes in high-ozone event frequency and duration from the principal component time series associated with each regional EOF.
5.043 On quantification of hydroxyl in chemistry-climate models.

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Abstract:

Hydroxyl (OH) radicals are in the heart of tropospheric air pollution chemistry. Yet no consensus is established between the atmospheric chemistry-transport model (CTM) simulated global mean abundance of OH and that estimated by proxy method using the decay rate of methyl chloroform (CH\textsubscript{3}CCl\textsubscript{3}) concentration. The CTM simulations probably overestimate global mean OH concentration by about 10% (Naik et al., 2013). The northern to southern hemispheric (NH/SH) ratio of OH simulated by the CTMs are in the range of 1.1 to 1.4, while the measurements and modelling of CH\textsubscript{3}CCl\textsubscript{3} inter-hemispheric differences suggest a parity in OH between the two hemispheres (Patra et al., 2014). I will review the present status of defining the global and hemispheric OH concentration using simulations of CH\textsubscript{3}CCl\textsubscript{3} by an online-transport and offline-chemistry model, i.e., JAMSTEC’s ACTM.

The bias in the NH/SH OH ratio has direct impact on the estimation of regional emissions by inverse modelling of the species that are oxidized in troposphere by reaction with OH, such as methane, carbon monoxide. As an example, methane emissions are estimated to be about 10% higher for USA and China if the NH/SH OH ratio is 1.26 compared to a case of NH-SH OH parity. Further, I will use the simulations of OH and related species from the ongoing CCMI model inter-comparison for testing the parity of NH/SH OH ratio. Various emission scenarios will be implemented for simulating the CH\textsubscript{3}CCl\textsubscript{3} decay rate for the CCMI modelled OH in the period 2000-2012. Finally, different sensitivity simulations of CCMI models and specially designed experiments by CHASER CTM will be explored for understanding feedback between OH concentrations and the chemistry of hydrocarbons.

Acknowledgements. The support of NOAA, AGAGE and HIPPO measurement teams, and availability of CCMI model simulations of OH and related species is greatly appreciated.
Ozone plays a key role in global climate change and atmospheric environment. Tropospheric ozone is not only a main greenhouse gas, but also a secondary pollutant. It has been known to the public because of extensive impacts on air quality, climate and human health. In this study, the inter-annual and seasonal variability of tropospheric ozone over East Asia were analyzed. The regional climate chemistry model (RegCM4+Chem) were evaluated and used to investigate the effects of East Asian Summer Monsoon (EASM) on ozone. Ozone variance analysis between strong and weak EASM proved that anomalies of EASM have a significant impact on spatial distribution of low layer ozone. Ozone was higher in the North during strong monsoon season and lower in the south during weak monsoon season. Ozone difference range between strong EASM and weak EASM was -6.5 to 6.9 ppb from May to August with the biggest difference in August. Average difference of 4 months ranged from -3 ppb to 3.8 ppb, accounting for -10% to 12.5% of the average ozone over the area. Process analysis indicated that advection and chemical reaction played the main role in the variation of ozone affected by EASM. Via wind and cloud, strength anomalies of EASM influenced transportation and chemistry formation of ozone and eventually lead to the differences of ozone spatial distribution at lower atmosphere.
Ocean-Atmosphere Exchange of Ammonia in the 21st Century and the Competing Effects of Temperature and Ocean Acidification.

Early Career Scientist

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Abstract:

Ammonia is the principal alkaline gas in the atmosphere. It therefore plays an important role in atmospheric chemistry, reacting with sulphuric and nitric acids to form ammonium aerosols, which serve as cloud condensation nuclei and negatively impact human health. Anthropogenic ammonia emissions are increasing rapidly in many areas of the world, and are expected to increase dramatically in the future due to the strong effect of temperature on the emission of ammonia. It is therefore of interest to understand the impact of increasing temperatures, atmospheric CO$_2$, and anthropogenic emissions on ocean-atmosphere exchange. Precise estimates of ocean-atmosphere ammonia exchange are unavailable due to the variability of fluxes and the difficulties in measuring them. A modelling approach is therefore required. Global ocean-atmosphere exchange of ammonia was simulated, applying an observationally-constrained physico-chemical model, for present-day and future scenarios, taking into account changes in temperature, terrestrial ammonia emissions, and ocean pH. Results show that ocean acidification has the largest effect, leading to a decrease in global ocean ammonia emissions from a range of 2.8 to 6.6 Tg-N/yr for the present day to a range of -1.1 to 2.3 Tg-N/yr for 2100 (RCP 8.5). Ongoing work includes incorporating this interactive ocean-atmosphere ammonia exchange scheme into the global atmospheric chemistry and aerosol model UKCA to explore resulting impacts on atmospheric composition and the global nitrogen cycle.
5.050 An uncertain future for lightning and the consequences for atmospheric composition and radiative forcing.

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Abstract:

Most studies to date suggest that lightning NO\textsubscript{X} emissions (LNO\textsubscript{X}) will increase under climate change. Our new analysis of the ACCMIP multi-model ensemble highlights the importance of this NO\textsubscript{X} source, which leads to about 6.5 times more ozone production than surface NO\textsubscript{X} sources. Furthermore, we find that most ACCMIP models use a lightning parametrisation based on cloud-top height, and produce a linear increase in LNO\textsubscript{X} of 0.44±0.05 TgN K\textsuperscript{-1} with respect to changes in global mean surface temperature. However, ACCMIP models using alternative lighting parametrisations suggest that increasing temperatures may yield much smaller changes or even reductions in LNO\textsubscript{X}. This suggests that the LNO\textsubscript{X} response to climate change is highly dependent on lightning parametrisation.

We have recently developed and evaluated a new, more physically-based lightning parametrisation using upward ice flux, which is closely linked to thunderstorm charging theory. To quantify the response of LNO\textsubscript{X} to future climate we have performed simulations with the UKCA climate-chemistry model using both the traditional cloud-top height and our new ice flux approach. The two parametrisations simulate opposing global responses of lightning to climate change. We predict an increase in annual lightning NO\textsubscript{X} emission of 0.44 TgN K\textsuperscript{-1} with the cloud-top height approach, similar to ACCMIP, but a decrease of 0.15 TgN K\textsuperscript{-1} with the upward ice flux approach.

The contrasting climate responses of lightning simulated with the two schemes results in substantially different effects on tropospheric ozone, OH and methane lifetime. These contrasting lightning responses lead to large differences in radiative forcing from ozone and methane, although the difference in net radiative forcing is relatively small. The dependence of the simulated climate response to the lightning parametrisation used in models constitutes a key uncertainty in projecting future tropospheric composition and its climate impacts, and this should be addressed in future chemistry-climate model studies.
Aerosol optical depth (AOD) is affected by the size, optical characteristics, and hygroscopicity of particles, confounding attempts to link remote sensing observations of AOD to measured or modeled aerosol mass concentrations. In situ airborne observations of aerosol optical, chemical, microphysical and hygroscopic properties were made in summer 2013 in the southeastern United States in the daytime when fair-weather cumulus clouds were present. These conditions are typical of midlatitude, moderately polluted airmasses. We use these observations to constrain a simple aerosol model that is used to test the sensitivity of AOD to the various measured parameters. As expected, the AOD was found to be most sensitive to aerosol mass concentration and to aerosol water content, which is controlled by aerosol hygroscopicity and the ambient relative humidity. However, AOD was also fairly sensitive to the mean particle diameter and the width of the size distribution. These parameters are often prescribed in global models that use simplified modal parameterizations to describe the aerosol, suggesting that the values chosen could substantially bias the calculated relationship between aerosol mass and optical extinction, AOD, and radiative forcing. Further efforts to systematically characterize the aerosol characteristics within different airmasses are needed to reduce both modeling and remote sensing uncertainties, and should lead to improved confidence in estimates of the direct aerosol radiative effect.
5.055 Long-term trends of global tropospheric column ozone.

Early Career Scientist

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Abstract:

Tropospheric ozone is a greenhouse gas and pollutant detrimental to human health and crop and ecosystem productivity. Since 1990 a large portion of the emissions of anthropogenic precursor species that react in the atmosphere to produce ozone have shifted from industrialized North America and Europe to Asia. This rapid shift, coupled with limited ozone monitoring globally and in these nations in particular, has left scientists with limited capability to answer the most basic questions: Is ozone continuing to decline in nations with strong emission controls? To what extent is ozone increasing in the developing world? In response to these questions this presentation will show results from IGAC’s Tropospheric Ozone Assessment Report, and will present the first multi-instrument comparison of long-term tropospheric column ozone trends using co-located observations from satellites, FTIR and Umkehr. The study is aimed at evaluating the chemistry-climate models participating in the CCMI and TF-HTAP experiments.
Parallel simulations of methane oxidation in UM-UKCA reveal hemispheric biases in climatological oxidant fields and methane concentrations.

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Abstract:

Methane is the second most important anthropogenic greenhouse gas after carbon dioxide and a major ozone precursor. Its relatively short lifetime allows any changes in the methane burden to have near immediate effects on global climate. However, large uncertainties exist in the estimates of methane sources and hence in the global methane budget.

The UM-UKCA has been modified to use both prescribed offline oxidant fields and interactively simulated oxidant fields in parallel. Thus, in one simulation methane oxidation can be treated as both a linear first order process, removing the methane feedback onto itself, and a fully interactive process. The linear oxidation scheme allows methane emissions to be tagged by source type and region. The analysis presented here focuses on the source-sink-balance of methane in the early 2000s.

We show that methane observations can be used to assess the performance of the model, using a consistent method for comparison between a standard OH climatology (Spivakovsky) and a chemistry climate model oxidant field and analyse their effects on methane lifetime. The UM-UKCA OH field (North-South gradient 1.31) compares well with the ACCMIP multi-model mean. Methane concentrations are low biased with respect to observations at all latitudes but with an acceptable hemispheric gradient. The methane lifetime with respect to tropospheric OH loss is at the lower end of the ACCMIP multi-model mean. In contrast, the Spivakovsky OH field (North-South gradient 1.01) gives good agreement with methane observations with a small high bias in the Northern Hemisphere resulting in too strong a hemispheric gradient. The lifetime agrees well with observational constraints. We will present an analysis of the footprints of the different source types and regions which allows us to perform detailed comparison to observations. We will show that these data allow a quantitative assessment of the speciated methane source strengths.
5.058 Quantifying and Reducing Uncertainty in Model Studies of Tropospheric Composition.

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Abstract:

Projections of future atmospheric composition change and its impacts on air quality and climate are heavily dependent on chemistry-climate models that allow us to investigate the effects of changing emissions and meteorology. These models are imperfect, as they rely on our understanding of the chemical, physical and dynamical processes governing atmospheric composition, on the approximations needed to represent these numerically, and on the limitations of the observations required to constrain them. Model intercomparison studies, like those initiated through HTAP, ACCMIP and CCMI, show substantial diversity in results that reflect these underlying uncertainties, but little progress has been made in identifying the weaknesses in process understanding or representation that might lead to improved models and to better scientific understanding. Observations provide a valuable constraint for models, but it is generally not possible to isolate and evaluate the effect of individual processes in the troposphere due to the interaction of processes occurring on similar spatial and temporal scales.

Model emulation and uncertainty analysis provide a new method of identifying and quantifying the main sources of uncertainty in current models. We apply a multi-variable perturbation approach to quantify the sensitivity of ozone and OH to important climate-relevant variables, poorly-characterized processes and uncertain anthropogenic emissions, using two independent global chemistry transport models, the FRSGC/UCI CTM and the GISS GCM. We show a clear sensitivity of tropospheric ozone to atmospheric humidity and precursor emissions which is similar for the models, but note large differences for methane lifetime, highlighting substantial differences in the sensitivity of OH to primary and secondary production. This approach allows us to identify key areas where model improvements are required while providing valuable new insight into the processes driving tropospheric composition change.
Abstract:

Future changes in tropospheric ozone (and its precursors), aerosols, greenhouse gases, and ozone depleting substances will reflect the complex interplay between changes in species' emissions, chemistry and transport. Here we present an analysis of tropospheric transport using idealized age tracers that probe different aspects of the transport circulation, based on simulations subject to present-day and future greenhouse gases and ozone depleting substances. A comparison between simulations of the Goddard Earth Observing System Chemistry Climate Model and the Whole Atmosphere Community Climate Model, wherein the large-scale flow is constrained to MERRA meteorology, reveals large (30%) differences in the mean transit time since air in the Southern Hemisphere last contacted the Northern Hemisphere (NH) midlatitude surface. This is interpreted largely in terms of differences between models' representations of convective transport, particularly over the oceans. Shifts in the location and strength of tropical convection and changes in isentropic transport out of the NH midlatitude surface layer are used to interpret future changes in transport over the twenty-first century. Comparisons with other models in the Chemistry Climate Modeling Initiative (CCMI) are used to further elucidate how intermodel differences in large-scale dynamics and convection translate to differences in the variability and long-term trends in transport.
5.062 Injection of iodine to the stratosphere and implications for future ozone depletion.

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Abstract:

There are still many uncertainties about the influence of iodine chemistry in the stratosphere, as the real amount of reactive iodine injected to this layer from the troposphere and the partitioning of iodine species are still unknown. In this work we report a new estimation of the injection of iodine into the stratosphere based on recently published (Saiz-Lopez et al., 2015) aircraft observations in the tropical tropopause layer (TORERO campaign) and a 3D global chemistry-climate model (CAM-Chem) that includes the most recent knowledge about iodine photochemistry. The results indicate that significant levels of total reactive iodine (0.25-0.7 pptv), between 2 and 5 times larger than the accepted upper limits, could be injected into the stratosphere via tropical convective outflow. At these iodine levels, modelled iodine catalytic cycles account for up to 30% of the contemporary ozone loss in the tropical lower stratosphere and can exert a stratospheric ozone depletion potential equivalent or even larger than that of very short-lived (VSL) bromocarbons. Therefore, in this presentation, we discuss the impact of iodine and VSL sources and chemistry on historical and future evolution of the stratospheric ozone layer.

5.063 Evaluating the Atmospheric Chemistry Implications of Climate Policies for Human Health in the U.S. and China.

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Abstract:

We use atmospheric modeling, coupled with economic modeling and health impacts analysis, to compare the health implications of carbon policies for the U.S. and China. Climate policies can have a variety of benefits for air quality, in particular the concentrations of health-damaging pollutants O$_3$ and PM$_{2.5}$ as well as toxic pollutants such as mercury. Meteorological changes that result from changing climate conditions (e.g. increased temperatures, stagnation) can affect the concentrations and distribution of pollutants; if future climate changes lead to increased pollution, mitigating climate change can have direct air pollution benefits. In addition, controlling CO$_2$ sources such as power plants and vehicles can lead to concomitant reductions in other pollutants such as SO$_2$, NOx, and Hg - so-called “co-benefits.” Here, we compare the magnitude of both direct benefits and co-benefits for carbon policies in the U.S. and China. In particular, we assess how policy design, stringency, and context affect the projected air pollution and health implications. We find that while U.S. co-benefits exhibit diminishing returns to projected increases in policy stringency, Chinese co-benefits increase with more aggressive climate goals. The magnitude of health-related co-benefits is sensitive to the choice of health impact function, with China-specific functions yielding substantially less co-benefit than those typically used in the U.S. We find that China’s direct benefit is more variable across polluted regions than those accrued in the U.S. In both cases, we see overall benefits but substantial regional variation, illustrating the benefits of coupled atmospheric-economic analysis.
Abstract:

Carbon Monoxide is a key component in tropospheric chemistry. It plays an important role by affecting the oxidative capacity through its loss with OH and being a precursor of tropospheric ozone. Both meteorological observations and multispectral retrievals of CO partial columns obtained from the MOPITT (Measurement of the Pollution in the Troposphere) instrument have been assimilated into the Community Atmosphere Model with Chemistry (CAM-CHEM), the atmospheric component of the Community Earth System Model (CESM). The assimilation is carried out using an Ensemble Adjustment Kalman Filter algorithm within the Data Assimilation Research Testbed (DART) package. We investigate the causes of the CO trends by analyzing chemical fluxes and CO emission tags spatio-temporal distribution and long-term trends. In particular, we discuss the assimilation results with regards to the oxidative capacity (i.e., OH distribution, methane lifetime) together with CO ad O₃ chemical production and losses. The results are compared to independent in-situ observations (e.g. MOZAIC-IAGOS aircraft observations, WDCGG surface measurements) as well as a CAM-Chem control run forced by MERRA reanalysis.
Estimation of sulfuryl fluoride emissions at regional and global scales by inverse modeling using AGAGE measurements.

Early Career Scientist

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Abstract:

Sulfuryl fluoride (SO$_2$F$_2$) is used increasingly as a fumigant to replace methyl bromide (CH$_3$Br), which was regulated under the Montreal Protocol (1986). Mühle et al. (2009) showed that SO$_2$F$_2$ had been accumulating in the global atmosphere with a growth rate of 5±1% per year from 1978 to 2007. They also demonstrated, using the 2D AGAGE box model, that SO$_2$F$_2$ has a total atmospheric lifetime of 36±11 years mainly driven by the oceanic uptake. In addition, the global warming potential of SO$_2$F$_2$ has been estimated to be ≈4780 for a 100-year time horizon (Papadimitriou et al., 2008), which is similar to the CFC-11 (CCl$_3$F) GWP. Thus it is a potent greenhouse gas and its emissions are expected to continue to increase in the future. Therefore, it is important that we improve our knowledge of past and current SO$_2$F$_2$ emissions and chemistry in the atmosphere. The focus of this work is to estimate SO$_2$F$_2$ surface emissions both at regional and global scales from 1978 to 2015. We use the 3D chemical transport model MOZART-4 with a 1.9° × 2.5° horizontal resolution to which we have added the main processes involved in SO$_2$F$_2$ atmospheric chemistry. We perform inverse modeling to improve the estimate of the SO$_2$F$_2$ emissions via the CELS (Combined Eulerian and Lagrangian Sensitivity) approach. This method uses the Eulerian MOZART-4 model and the lagrangian NAME model together with the AGAGE (Advanced Global Atmospheric Gases Experiment) measurements at Mace Head (Ireland), Trinidad Head (California), Gosan (South Korea), Shandianzi (China), Cape Grim (Australia), Cape Matatula (Samoa), Ragged Point (Barbados), Jungfraujoch (Switzerland), Monte Cimone (Italy) and Ny-Alesund (Svalbard), along with air archives going back to 1978. Results will be compared to the global industrial estimates and provide a much better understanding of the SO$_2$F$_2$ emissions at regional and global scales over the last decade.
5.067 Sources and impacts of short-lived OVOC in the remote tropical marine troposphere.

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Abstract:

The tropical free troposphere is a key atmospheric environment to understand chemistry-climate interactions. About 75% of the tropospheric ozone (O$_3$) and methane (CH$_4$) loss occurs at tropical latitudes. There is a particular need to understand the emissions and chemistry of tropospheric halogens and organic carbon in pristine remote environments, because they influence the reactive chemical removal pathways of climate active gases, and can modify aerosols that affect Earth’s radiation balance. About 45 per cent of the variance of aerosol forcing since about 1750 arises from uncertainties in natural emissions.

Recently, field measurements over the remote tropical Eastern Pacific ocean were conducted using aircraft and ships during the Tropical Ocean tRoposphere Exchange of Reactive halogen species and Oxygenated VOC (TORERO) field campaign (Jan/Feb 2012). We have shown that tropospheric halogen chemistry has a larger capacity to destroy O$_3$ and oxidize atmospheric mercury than previously recognized. Halogen chemistry is currently missing in most global and climate models, and helps explain the low O$_3$ levels in preindustrial times. Marine emissions of organic carbon include VOCs (isoprene, monoterpenes), primary organic aerosol (POA), dimethylsulfate (DMS), and oxygenated hydrocarbons (OVOC, i.e., acetaldehyde, acetone, methanol, aliphatic aldehydes, and
glyoxal). Notably, the sum of acetaldehyde and glyoxal emissions accounts for >60% of the overall marine organic carbon emissions. However, the source mechanism for these OVOC is currently poorly understood.

The experimental results from TORERO are summarized, and used in conjunction with models to quantify the atmospheric implications of the measured OVOC and halogen radical abundances for O$_3$ loss, HO$_X$ radical sources, oxidative capacity, and bromine radical sinks. Our results suggest that ocean impacts on the upper free troposphere are underestimated, and that the chemistry of tropospheric halogens and OVOC is more closely coupled than previously believed. Our understanding of the biogeochemical cycle of marine organic carbon is incomplete.
5.068 Multi-model comparison of marine boundary layer O3 in HTAP2 simulations with cargo ship observations in Asia-Pacific.

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Abstract:

We evaluated surface ozone (O3) simulated by global and hemispheric chemical transport models (CTM) participating in the Hemispheric Transport of Air Pollution (HTAP) Phase 2 by comparing with observations obtained on commercial cargo ships plying between Japan and Southeast Asian or Oceanian regions. Reproducibility of seasonal cycles of trace gases is the most common test for an evaluation of CTMs and an important requirement demonstrating model performance. The long-term observations from the cargo ships for about 10 years provide climatological seasonal variations and latitudinal gradients of O3 in the marine boundary layer across a wide range of latitudes (40°S–35°N) on the western Pacific, East China, and South China Seas. At the northern midlatitudes, surface O3 concentrations show a seasonal maximum during spring and minimum in summer, reflecting the seasonal cycle of the Asian monsoon. The CTMs generally reproduced the springtime peak of O3 in April-May. However, the CTMs tended to overestimate the observed summer minimum in O3 at 20–30°N on the Japan–Southeast Asia (mean bias of 12–14 ppbv in June–August) and the Japan–Oceania
routes. In addition, we found that the CTM results exhibited a similar positive bias of surface O$_3$ at the remote sites located in the same latitudinal regions (e.g., Okinawa and Ogasawara). The overestimates across wide areas of the Asian seas suggest that chemical loss and/or deposition processes of marine boundary layer O$_3$ are underestimated by the CTMs. The positive bias in this season may contribute to the overestimation of modeled surface O$_3$ around Japan, which was reported by the model intercomparison studies, because southerly winds prevail around the rim of the North Pacific High during summer. We have revealed the spatial structure of the summertime O$_3$ overestimation in the Asian region in many CTMs, which is useful to clarify the causes of this problem.
5.070 The Stratospheric Contribution to Tropospheric Ozone Variability and Trends.

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Abstract:

Chemistry-climate models predict an intensification of the stratospheric circulation over the next century in response to greenhouse gas forcing, with uncertain consequences for tropospheric ozone and climate. We examine the role of the stratosphere in present-day tropospheric ozone variability and trends as measured by the Tropospheric Emission Spectrometer (TES) and Microwave Limb Sounder (MLS) onboard NASA’s Aura satellite. Our previous work has shown that interannual variability in the stratospheric circulation of ±40% leads to changes of ±25% in northern midlatitude lower stratospheric ozone and ±2% in northern midlatitude tropospheric ozone. Here we examine in more detail the drivers of stratospheric circulation variability and the subsequent ozone response using the Whole Atmosphere Chemistry-Climate Model (WACCM) in order to better understand the relationship between interannual and long-term changes in circulation and ozone. We find that both the stratospheric Quasi-Biennial Oscillation (QBO) and El Nino / Southern Oscillation (ENSO) drive stratospheric circulation changes but that both the circulation changes and ozone response depend critically on the timing of QBO and ENSO relative to one another and to the seasonal cycle. We also examine the role of stratospheric variability in tropospheric ozone trends over the past decade and show that
this variability can confound quantification of emissions-driven changes. We discuss the implications of our work in terms of reducing uncertainties in long-term projections of tropospheric ozone.
5.071 Impacts of precipitation patterns on the wet deposition and lifetime of aerosols.

Early Career Scientist

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Abstract:

Wet deposition is a major removal process for many air pollutants including aerosols and other soluble species. Global climate change can significantly affect the precipitation amount and patterns which have important implications for the wet deposition process. As part of a study examining the long-term changes in extreme air pollution meteorology based on the global observational data for the period of 1951-2010, we analyze the long-term trends in the precipitation characteristics including precipitation amount, frequency, intensity, duration, type (rain or snow) and diurnal variation. Some significant trends have been identified. For example, we find decreasing precipitation frequency over Europe, North Africa, Asia and South America. We then conduct multiple sensitivity simulations with the GEOS-Chem chemical transport model to quantify the impacts on aerosols from the changes in these precipitation characteristics. Our results show that the atmospheric aerosols’ lifetime against wet deposition is most sensitive to precipitation frequency. This implies that the decreasing precipitation frequency would lead to longer atmospheric lifetime of aerosols, even the total precipitation amount remains unchanged.
Evaluation of the co-benefits of low carbon policies on residential sector in Asian region for air pollution abatement.

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Abstract:
In many Asian countries, the GHG and Short lived Climate Pollutants(SLCPs) emission from residential sector accounts for large percentage. Many countermeasures are proposed for GHG reduction, and they also can reduce an emission of SLCP at the same time. In this study, the emission of SLCPs from residential sector in Asian countries and its environmental fate were investigated. First, the current and future air pollutants emissions from residential sector in Asian countries were estimated using bottom-up type energy enduse technology selection model. Energy balance statistics and domestic statistics for energy consumption and energy technology used in residential sector were used as an input data. Then, the regional air quality simulation were carried out to investigate the contribution to the ambient concentration of SLCP using chemical transport model, CMAQ under the current meteorological condition for one year. We estimated the contribution of emission from residential sector to the concentration of PM2.5. Also, the effect of several reduction countermeasures for residential sector were estimated and reduction effect of ambient concentration were calculated. As a result, contribution of the residential sector to PM2.5 concentration in many part of Asia were calculated as 30-60%. Large seasonal variation were shown in high-latitude region. Finally, the health impact were estimated using similar method of Global Burden of Disease (WHO). We estimated the reduction of premature death due to residential emission under the each countermeasures.
Air pollutants in the Arctic have impacts on climate change, ecosystems, regional air quality, and human health. Rapid changes to and complex interactions within the Arctic environment due to global warming and socio-economic drivers mean that there is an urgent requirement to improve understanding of sources of Arctic air pollutants. Changes in atmospheric aerosol particles and tropospheric ozone have likely contributed substantially to rapid warming of the Arctic over recent decades. Increased accessibility due to reducing sea-ice is leading to increased local pollutant emissions from activities such as oil and gas extraction or shipping. Local Arctic communities are already being affected by sources of air pollution, which may continue to grow as economic and industrial development continues in the region. It is crucial to improve quantification of the relative contributions of different pollutant sources to provide a sound scientific basis for sustainable solutions and adaptive strategies. Deficiencies in predictive capability and a lack of observations at high latitudes present major challenges to advancing this understanding, and to making credible near- and long-term projections of Arctic environmental change.

This poster will describe a new international initiative - **Air Pollution in the Arctic: Climate, Environment and Societies (PACES)** (see www.igacprojects.org/PACES), focused on building capacity for future research on Arctic air pollution. We will present the key scientific motivating factors behind the establishment of PACES, and our plans for addressing current uncertainties and deficiencies in our understanding of sources, processing and fate of air pollutants in the Arctic, and their impacts on human health, ecosystems and climate. These plans include development of strategies for targeted field observations addressing key processes, improving predictive capability, improving observational capacity in the Arctic, particularly in Russia, and in the vertical, and via
establishment of collaborations between physical scientists, social scientists and local residents.
5.077 An atmospheric definition of the equator and its implications for atmospheric chemistry and climate.

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Abstract:

Earth’s northern hemisphere contains higher concentrations of many greenhouse gases and air pollutants than the southern hemisphere due to human activity and the partial isolation of the atmosphere’s northern and southern hemispheres. While sharp composition gradients have been observed in the tropics for decades, the boundary between the atmospheric hemispheres has not been rigorously defined. After showing that the intertropical convergence zone (ITCZ) and other past definitions are not adequate, we suggest defining the atmospheric equator as the boundary where air originates equally from the northern and southern extra-tropics. We use a 3-D atmospheric transport model to identify the location and spatio-temporal structure of the atmospheric equator. This definition coincides with observed concentration gradients and the mean position of the ITCZ, when it is well developed over the oceans, and also works in regions where the ITCZ is ill defined. On average the southern hemisphere is larger, with large seasonal and synoptic variability. The asymmetry of the atmospheric hemispheres affects how simple mass balance models are constructed and evaluated against observations. For example, erroneously assuming equal hemisphere sizes overestimates the rate of interhemispheric transport implied by SF$_6$ observations. We also show that tropospheric OH concentrations and CH$_4$ loss rates in 3-D chemistry models are substantially different when averaged over atmospheric vs. geographic hemispheres. Methyl chloroform observations imply that CH$_4$ loss in the southern atmospheric hemisphere is equal to or faster than in the north, but for decades models have predicted much faster CH$_4$ loss in the northern hemisphere. Diagnosing the models consistently with the observational constraints at the atmospheric equator reveals that modeled north-south differences of CH$_4$ loss are actually consistent with observations within their uncertainties. This result supports our ability to simulate a key atmospheric oxidant, tropospheric OH, and its consequences for air quality and climate.
Secondary organic aerosol (SOA) plays a significant role in Earth system by altering its radiative balance. Here, we use an earth system model coupled with an explicit SOA formation module to estimate the response of SOA concentrations to changes in climate, anthropogenic emissions and human land use in the future. We find that climate change is the major driver for SOA change under the RCP8.5 future scenario. Climate change increases the isoprene emission rate by 18% with the effect of temperature increases outweighing those of the CO\textsubscript{2} inhibition effect. As a result, SOA is increased by 25%. We also separately evaluate the effects of changes in anthropogenic emissions and land use change and find that these decrease SOA. We will present these results and contrast them with results from the existing literature.
5.081 Climate impact of emissions of short-lived climate forcers.

Early Career Scientist

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Abstract:

There is a growing interest in the potential for reducing short-lived climate forcers (SLCFs) to slow Arctic and global warming and at the same time improve air quality. In this study we have systematically quantified the Arctic climate impact of regional SLCFs emissions, taking into account black carbon, sulphur dioxide, nitrogen oxides, volatile organic compounds, organic carbon and tropospheric ozone, and their transport processes and transformations in the atmosphere. Using several chemical transport models we perform detailed radiative forcing calculations from emissions of these species from different sectors and regions. To estimate the Arctic surface temperature we apply regional climate sensitivities, the temperature response per unit of radiative forcing for each SLCF in four broad latitude bands. We find that the largest Arctic warming source is from emissions within the Asian nations owing to the large absolute amount of emissions. However, the Arctic is most sensitive, per unit mass emitted, to SLCFs emissions from a small number of activities within the Arctic nations themselves. A stringent, but technically feasible mitigation scenario for SLCFs, phased in from 2015 to 2030, could cut warming by 0.2 (±0.17) K in 2050. To extend the process-based understanding of the emissions-to-response approach we run a fully coupled climate model with emission perturbations of black carbon and sulphur dioxide over Europe, USA, South Asia and East Asia. We find significant changes in temperature and precipitation over many regions in the Northern Hemisphere (compared to a 200 year control run). We see similar response patterns in the Northern Hemisphere (locally and remotely) from emissions perturbations from very different regions (USA vs. East Asia).
Evaluating surface fluxes of methane using total column data requires models to accurately account for the transport and chemistry of methane in the free troposphere and stratosphere, thus reducing sensitivity to the underlying fluxes. Vertical profiles of methane have increased sensitivity to surface fluxes because lower tropospheric methane is more sensitive to surface fluxes than a total column and quantifying free tropospheric CH$_4$ concentrations helps to evaluate the impact of transport and chemistry uncertainties on estimated surface fluxes. Here we demonstrate new estimates of lower tropospheric CH$_4$ concentrations through the combination of free tropospheric methane measurements from the Aura Tropospheric Emission Spectrometer (TES) and XCH$_4$ (dry-mole air fraction of methane) from the Greenhouse Gases Observing Satellite Thermal And Near Infrared for Carbon Observations (GOSAT TANSO, herein GOSAT for brevity).

The calculated precision of these estimates ranges from 10 to 30 ppb for a monthly average on a 4x5 latitude / longitude degree grid making these data suitable for evaluating lower-tropospheric methane concentrations. Smoothing error is approximately 10 ppb or less. Comparisons between these data and the GEOS-Chem model demonstrate that these lower-tropospheric CH$_4$ estimates can resolve enhanced concentrations over flux regions that are challenging to resolve with total column measurements. We also use the GEOS-Chem model and surface measurements in background regions across a range of latitudes to determine that these lower-tropospheric estimates are biased low by approximately 65 ppb, with an accuracy of approximately 6 ppb (after removal of the bias) and an actual precision of approximately 30 ppb. This 6 ppb accuracy is consistent with the accuracy of TES and GOSAT methane retrievals.
6.003 Long-term measurements of atmospheric inorganic gaseous species at Cape Point, South Africa.

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Abstract:

The Cape Point Global Atmosphere Watch (CPT GAW) station is globally considered to be an important atmospheric monitoring site due to its position at the south-western tip of Africa. CPT GAW is a baseline station measuring long-term trends under background conditions, which is predominantly impacted by clean maritime air masses. However, it is also impacted by local sources of atmospheric pollutants, which include the greater Cape Town conurbation and other industrial activities in this region. CPT GAW is also part of the southern African IGAC DEBITS Africa (IDAF) network. Long-term passive sampler measurements of atmospheric inorganic gaseous species, i.e. sulphur dioxide (SO₂), nitrogen dioxide (NO₂), ammonia (NH₃), nitric acid (HNO₃) and ozone (O₃) conducted at CPT GAW from 1995 to 2015 were assessed in order to establish seasonal patterns, inter-annual variability and long-term trends of these compounds. Distinct seasonal patterns were observed for SO₂, NO₂ and O₃, with concentrations of these species peaking during the wet winter/early spring months (June-September). The seasonality was attributed to an increase in long-range transport of pollutant species from the industrialised interior of South Africa and larger impacts of air masses passing over the Cape Town conurbation. Higher NO₂ concentrations were also attributed to increased microbial activity occurring with the onset of the wet season. The inter-annual average concentrations of SO₂ and NO₂ revealed a steady decrease up until 2002, after which an increase in annual average concentrations was observed. These increases could partially be attributed to economic growth and the increasing population in South Africa. In an effort to substantiate long-term trends, the dataset will also be subjected to a multilinear regression model in order to establish the influence of changes in regional and global meteorological patterns on concentrations of these inorganic gaseous species, which will be reported at the IGAC 2016 conference.
6.004 Wintertime Reactive Nitrogen Chemistry During the 2015 WINTER Aircraft Campaign.

Early Career Scientist

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Abstract:

Tropospheric ozone (O$_3$) is a potent greenhouse gas that degrades regional air quality. Anthropogenic NO$_x$ emissions and chemistry are an important regional O$_3$ source during midlatitude summer, but may destroy O$_3$ during winter. The large majority of previous field studies have focused on understanding reactive NO$_x$-O$_3$ relationships under summertime conditions. However, there remain outstanding scientific questions regarding the influence of NO$_x$ on wintertime O$_3$, in part because of the relevant atmospheric chemistry that occurs largely at night, including N$_2$O$_5$ multiphase processes. The winter tropospheric O$_3$ budget depends critically on N$_2$O$_5$ production and the efficiency of its subsequent reaction on aerosols to nitric acid (HNO$_3$) or nitryl chloride (ClNO$_2$). These processes are highly uncertain due in part to the limited database for wintertime atmospheric chemical measurements.

The Wintertime Investigation of Transport, Emissions, and Reactivity (WINTER) campaign conducted 13 research flights over the eastern US in February and March 2015. A wide variety of environments were sampled during day and overnight flights over continental and marine environments. Reactive nitrogen measurements were collected with a cavity ring down spectrometer, while HNO$_3$ and ClNO$_2$ were measured with chemical ionization mass spectrometry. Initial analysis of these data has shown that wintertime N$_2$O$_5$ mechanisms and reactive nitrogen partitioning are highly variable. Presented here is further analysis aimed at quantifying the observed N$_2$O$_5$ aerosol reactive uptake coefficient as a function of different ambient conditions observed during WINTER (e.g. temperature, relative humidity, gas vs. particle phase nitrate and chloride). In addition, a chemical box model is used to assess the chemical and/or environmental factors that influence reactive nitrogen partitioning.
Using Self-Organizing Map (SOM) Clusters to Create Ozone Profile Variability and Pollution

Early Career Scientist

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Abstract:
A typical way to formulate O₃ climatologies is to average ozonesonde profiles on a monthly or seasonal basis, either for specific regions or zonally. However, the variability we are trying to capture in a tropospheric climatology reflects meteorological conditions that may not be confined to a particular season. We demonstrate the advantages of using a statistical clustering technique, self-organizing maps (SOM), over simple averaging, through analysis of more than 4500 sonde profiles taken from the long-term US sites at Boulder, CO; Huntsville, AL; Trinidad Head, CA; and Wallops Island, VA. We apply SOM to O₃ mixing ratios from surface to 12 km (above MSL). At all four sites, profiles in SOM clusters exhibit similar tropopause height, 500 hPa height and temperature, and total and tropospheric column O₃. When profiles from each SOM cluster are compared to monthly O₃ means, near-tropopause O₃ in three of the clusters is double (over +100 ppbv) the climatological O₃ mixing ratio. Large mid-tropospheric deviations from monthly means (-6 ppbv, +7 – 10 ppbv O₃ at 6 km) are found in two highly-populated clusters with a combined 36–39% of profiles. The latter clusters represent both distinctly polluted (summer) and clean (fall/winter, high tropopause) O₃ profiles. In the case of Trinidad Head, SOM clusters of O₃ profile data from the lower troposphere can discriminate background vs polluted O₃ and the conditions linked with each. Two of nine O₃ clusters exhibit thin layers of high O₃, typically between 1 and 4 km, that reside above a subsidence inversion. Ozone in these clusters is upwind of high-altitude surface O₃ monitors at inland locations. Days corresponding to the high O₃ clusters exhibit hourly surface O₃ anomalies at the surface sites of +5 –10 ppbv compared to a climatology; the anomalies can last up to four days.
Seasonal variation of nitro-aromatic constituents in size segregated water soluble organic aerosols.

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Abstract:

Organic carbon represents a substantial fraction of atmospheric aerosol; hence, it is a very important contributor to the Earth’s climate, defining air quality and adverse health effects. It has profound influence on atmospheric chemistry. Besides altering the hygroscopic properties, surface tension and effective density of aerosol, water soluble organic carbon (WSOC) exhibits also light-absorbing properties (“brown carbon” - BrC). The colored light-absorbing compounds are very complex, and our understanding of the relationship between the chemical composition and optical properties of BrC remains limited.

Nitro-aromatic compounds (NAC) absorb UV/VIS light, and hence, could affect the radiative balance. Studies on the composition of BrC in humic-like substances isolated from biomass-burning aerosols have demonstrated the presence of yellow-colored nitrocatechols\(^2\). In addition to 4-nitrocatechol as the prevailing NAC, several other NAC have been identified and quantified in PM\(_{10}\)\(^3\). Although knowledge of the size distribution of aerosol components provides important information about their sources, formation and aging mechanisms in the atmosphere, the size distribution of NAC and their quantitative contribution to WSOC is completely unknown.

A comprehensive study on seasonal aerosol size-segregated WSOC, with an emphasis on its NAC (4-nitrocatechol; methyl nitrocatechols, (di)nitrophenols, methyl nitrophenols, nitrosalicylic acids, (di)nitroguaiacols) through investigation of their molecular level speciation and quantification will be presented. Size-segregated aerosols (size range: 0.038-15.6 \(\mu\)m) were collected in the urban background environment of Ljubljana, Slovenia. Chemical characterization of WSOC included measurements of their light absorption properties by UV-VIS spectrometry and molecular level determination of target NAC by applying LC-MS. The obtained data were correlated with those for levoglucosan as a tracer for biomass burning emissions as well as with aerosol mass, total carbon (TC), and carbon content in WSOC.

References:

6.009 Design of a novel aircraft open-path cavity ringdown spectrometer.

Early Career Scientist

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Abstract:

Aerosols and their effect on the radiative properties of clouds contribute one of the largest sources of uncertainty to the Earth’s energy budget. Many current global assessments, of atmospheric aerosol radiative forcing rely heavily on remote sensing observation; therefore, *in situ* aircraft and ground-based measurements are essential for validation of remote sensing measurements. Cavity ringdown spectrometers (CRD) measure aerosol extinction and are commonly used to validate remote sensing...
observations. These instruments have been deployed on aircraft based platforms over the years thus providing the opportunity to measure these properties over large areas in various conditions. However, deployment of the CRD on an aircraft platform has drawbacks. Typically, aircraft based CRDs draw sampled aerosol into a cabin based instrument through long lengths of tubing. This limits the ability of the instrument to measure:

1) Course mode aerosols (e.g. dust)
2) Aerosols at high relative humidity (above 90%)

Here we describe the design of a novel aircraft based open path CRD. The open path CRD is intended to be mounted external to the cabin and has no sample tubing for aerosol delivery, thus measuring optical properties of all aerosol at the ambient conditions. However, the design of an open path CRD for operation on a wing-mounted aircraft platform has certain design complexities. The instrument’s special design features include 2 CRD channels, 2 airfoils around the open Path CRD and a configuration which could be easily aligned and rigid at the same time. This novel implementation of cavity ringdown spectroscopy will provide a better assessment of the accuracy of remote sensing satellite measurements.
6.011 Enabling the use of low cost air quality sensors for atmospheric chemistry research.

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Abstract:

Low cost air quality sensors have been widely publicized, in principle offering increased information on the spatial and temporal variability of important chemical species and potentially a paradigm shift in air pollution monitoring. Despite multiple ‘citizen science’ applications of air quality sensors, uptake of these technologies by the ‘academic’ atmospheric science community has been slow due to concerns over selectivity, sensitivity, accuracy, precision and temporal response. We report a range of laboratory and field studies of commonly-used sensors. In the laboratory we undertook a multicomponent calibration, exposing sensors to a mixture of gases at concentrations typical of an urban environment. We show a number of cross-interferences between the stated sensor analyte and the other species in the calibration mix, several of which would dominate sensor signals under typical city conditions. In field tests we ran twenty identical commercial sensor packages alongside standard regulatory instruments in a suburban environment over a period of three weeks. This experiment highlighted significant issues with sensor-to-sensor variability, as well as with signal response to target molecules for certain sensors.

We conclude that there are significant hurdles to be overcome if the current generation of low-cost air quality sensors are to be used for research or regulatory purposes. However, our results also indicate that there may be routes forward. We show that for some sensors, the median signal from an ensemble of sensors provides a significantly better estimate of the target pollutant concentration than any sensor. We also show that although not necessarily orthogonal, different sensors measure different properties of the atmosphere. Thus an instrument composed of an ensemble of multiple different sensors coupled to an appropriate calibration methodology and statistical model may allow the creation of low cost sensor systems for measuring the concentration of some atmospheric pollutants.
6.014 Insight into the Global Distribution of Ground-level Fine Particulate Matter from Satellite Remote Sensing and from the SPARTAN Aerosol Network.

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Abstract:

Fine particulate matter (PM$_{2.5}$) is believed to be the leading environmentally-related risk factor for premature mortality worldwide. However, ground-level monitoring remains sparse in many regions of the world. Satellite remote sensing from multiple platforms (e.g. MODIS, MISR, SeaWiFS, CALIOP) and algorithms (e.g. Dark Target, Deep Blue, MAIAC) offers a global data source to address this issue. Global numerical modeling plays a critical role in relating these observations to ground-level concentrations. The resultant satellite-based estimates of PM$_{2.5}$ indicate dramatic variation around the world, with implications for global public health. A new ground-based aerosol network (SPARTAN) offers valuable measurements about PM$_{2.5}$ in globally dispersed urban areas across more than 10 countries to evaluate and improve satellite-based PM$_{2.5}$ estimates, and to provide near-surface chemical and microphysical detail unobtainable from space. These SPARTAN measurements offer insight into the global mass, optical properties, chemical composition, and trace metals in PM$_{2.5}$. This presentation will highlight recent advances in combining satellite remote sensing, global modeling, and ground-based measurements of atmospheric aerosols to improve understanding of global population exposure to PM$_{2.5}$.

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Abstract:

Tropospheric ozone is a greenhouse gas and pollutant detrimental to human health and crop and ecosystem productivity. Since 1990 a large portion of the anthropogenic emissions that react in the atmosphere to produce ozone has shifted from North America and Europe to Asia. This rapid shift, coupled with limited ozone monitoring in developing nations, has left scientists unable to answer the most basic questions: Which regions of the world have the greatest human and plant exposure to ozone pollution? Is ozone continuing to decline in nations with strong emissions controls? To what extent is ozone increasing in the developing world? How can the atmospheric sciences community facilitate access to the ozone metrics necessary for quantifying ozone’s impact on human health and crop/ecosystem productivity? In response, IGAC initiated the Tropospheric Ozone Assessment Report (TOAR), to provide the research community with an up-to-date scientific assessment of tropospheric ozone’s global distribution and trends from the surface to the tropopause. TOAR has also built the world’s largest database of surface ozone observations and is generating ozone exposure and dose metrics at thousands of measurement sites around the world, freely accessible for research on the global-scale impact of ozone on climate, human health and crop/ecosystem productivity. This talk will present the latest findings from TOAR revealing the regions of the world with extreme and worsening levels of surface ozone pollution. From a broader perspective, recent satellite data show changes in the tropospheric ozone burden which will be assessed in light of current emissions inventories, satellite detected tropospheric NO2 and model analyses of the equatorward shift of ozone precursor emissions. Finally, the recent evolution of global ozone trends will be compared to the ACCMIP 2030 projections to gauge the expectation that the global tropospheric ozone burden will increase under business as usual emissions increases.
6.027 A First Look at the Global Distribution of Newly Formed Particles from the NASA Atmospheric Tomography Mission (Atom).

Early Career Scientist

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Abstract:

Aerosols affect climate by directly scattering sunlight and affecting cloud properties. Over half of global cloud condensation nuclei (CCN) come from particles formed in-situ from the gas phase rather than directly emitted particles. Particle formation mechanisms are poorly understood, and different mechanisms lead to different spatial distributions of particles. The probability of newly formed particles becoming CCN varies spatially e.g. with pre-existing condensation, so knowing the spatial distribution of nucleated particles is important in assessing their contribution to radiative forcing.

Measuring the global distribution of aerosols would allow evaluation of different particle formation mechanisms in global models. Remote areas are under-sampled but could provide the most direct evidence to distinguish between these mechanisms. Furthermore, newly formed particles may contribute more to CCN concentrations and direct effects in environments with low direct anthropogenic emissions. Current satellite sensors cannot directly measure particles <0.05μm. Previous airborne studies evaluating aerosol size distributions have tended to amalgamate data from separate campaigns, which often use differing methodologies, be regional in scale, or focus on specific phenomena.

A contiguous global dataset of nanoparticle size distributions, with coverage of remote areas, is needed to better constrain climate models and improve our understanding of new particle formation and its effects on climate. Here we present aerosol size distributions from the first of four around-the-world flights of the NASA Atmospheric
Tomography mission (ATom). The flights scanned the atmosphere in continuous ascents and descents (0.2-12km) over the Pacific and Atlantic with near-pole-to-pole coverage in July and August 2016. We report the first ultrafine aerosol size distributions (5-60nm) from ATom, measured with a suite of fast-response instruments, identifying large-scale spatial features from the lower to the upper troposphere, spanning remote polar, temperate, and tropical regions across both ocean basins.
6.028 Comparison of VOCs and their roles in ozone formation at a polluted site and a clean site in southern China.

Early Career Scientist

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Abstract:

Volatile organic compounds (VOCs) are important constituents in the lower troposphere, playing key roles in the formation of ozone (O_3) and secondary organic aerosols (SOA). Although high ground-level O_3 concentration has become a severe problem in both urban and rural areas in southern China, partly due to its hot weather, very limited information is known about the spacial difference of O_3 formation mechanisms in this region. More than fifteen kinds ambient VOCs were measured by a proton transfer reaction-mass spectrometer (PTR-MS) both at an urban site in Shenzhen (SZ) and a regional background site on Mount Wuzhi (WZ) in spring, in order to compare VOCs composition and contribution to the formation of O_3 at these two very different sites. The average measured total VOCs mixing ratio was 26.2 ppbv in SZ, which was far higher than that in WZ (8.2 ppbv). Oxygenated VOCs (OVOCs, 67%) and aromatics (30%) contributed the mostly in SZ, while OVOCs (75%) and biogenic VOCs (BVOC, 15%) were the most abundant species in WZ, where aromatics only contributed 8%. The emission ratios of toluene and C8 aromatics to carbon monoxide (CO) in SZ were about ten times larger than those in WZ, indicating very strong anthropogenic emissions of VOCs, especially aromatics in SZ. The photochemical reactivity analysis showed that aromatics and OVOCs
together consisted of around 90% of the ozone formation potential (OFP) among the measured VOCs in SZ, while BVOCs and OVOCs ranked the two largest species for the OFP in WZ. We also found the formation of O$_3$ was limited by VOCs in SZ, while it was more sensitive to nitrogen oxides (NOx) in WZ. These comparisons indicated that the O$_3$ formation mechanisms could be quite different in urban and rural areas in southern China, importantly due to the different VOCs sources and reactivity.
Long-term trends of tropospheric ozone precursors at the Czech EMEP and ACTRIS station Kosetice.

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Abstract:

Measurements of tropospheric ozone precursors (volatile organic compounds and nitrogen oxides) at the Kosetice Observatory (Czech Republic) has been implemented since 1993 within EMEP and later ACTRIS (after 2011) monitoring networks. The Observatory (49°35´N, 15°05´E, 534 m a.s.l.), operated by the Czech Hydrometeorological Institute (CHMI), was established in 1988 as a station specialized in air quality monitoring at the background scale. The nonparametric Mann-Kendall method was used for statistical evaluation of volatile organic compounds (VOCs) and nitrogen oxides (NOx) trends significance. In the Czech Republic, NOx emissions dropped by 60% in the period 1990-2012 and by 35% after 2000. The figures for VOCs were 68% in the period 1990-2012 and 38% after 2000. Very significant downward trend was found for almost all of measured VOCs with an exception of isoprene, which is controlled first of all by natural conditions and shows different patterns as other VOCs. On contrary, no trend was found by evaluation of NOx concentrations in the atmosphere, despite quite significant drop of the nitrogen emissions. The results from the Kosetice Observatory are in very good correspondence with the trends at similarly located stations in the neighbouring countries (Austria, Germany). The reasons are probably the significant changes in the structure of nitrogen emissions. The total NOx emissions are negatively affected by a considerable increase in emissions from mobile sources. The share of NOx emissions from mobile sources increased from 27.0% in 1990 to 55% by nowadays. The above mentioned trends of tropospheric ozone precursors were reflected in slightly significant downward trend of mean annual tropospheric ozone concentrations. More importantly, the number of episodes with target value for human health exceedances dropped significantly during the period 1993-2015.
Abstract:

The air pollution in Yangtze River delta (YRD), the largest economic region in China, threatens the health of the habitants in this region. Understanding the vertical distributions of the aerosols and the precursor trace gases in the boundary layer is very important to identify the dominating sources. However so far, there are few studies to reveal the vertical distributions from long-term field measurements. Recently some studies use satellite instruments and chemical transport models to identify and quantify the emissions of the pollutants in this region. However there are only few studies to verify their results and quantify the associated uncertainties. From 2011 to 2014 a MAX-DOAS instrument operated in Wuxi, China (belonging to YRD) is used to characterize the vertical
distributions of aerosols, NO$_2$, SO$_2$ and HCHO using our own profile retrieval algorithm. The data are used to characterize the seasonal, diurnal, and weekly variations of the species and to investigate the correlations between the different species. The vertical distributions derived from MAX-DOAS are also used to validate tropospheric NO$_2$, SO$_2$ and HCHO data from OMI and GOME-2. We find that the a-priori profiles in the satellite AMF calculations and the cloud shielding effect are the dominant error sources. MAX-DOAS results are also compared to hourly, daily and monthly averaged data from IMAGES, CHIMERE and Lotos-Euros models. Here considerable differences of the vertical distributions and diurnal cycles are found. The consistency depends strongly on the emission inventory used in the model simulations.

Early Career Scientist

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Abstract:

We measured the concentration, δ¹³C and δD of atmospheric CH₄ at Churchill (58°44’N, 93°50’W) in the northern part of Hudson Bay Lowlands (HBL), Canada during 2007–2014. Compared to the observational results at northern high-latitude background station, Ny-Ålesund, Svalbard (78°55’N, 11°56’E), the CH₄ concentration is generally higher and δ¹³C and δD are lower, reflecting the influence of regional biogenic CH₄ sources. A clear seasonal cycle is observable for both the CH₄ concentration and δ¹³C, with the respective maximum (minimum) values in January–February (June) and May (October). δD also shows the seasonal cycle, but it is not so clear as those for the CH₄ concentration and δ¹³C. The seasonal phases of the three variables are earlier at Churchill than at Ny-Ålesund by up to one month, due to the difference in seasonally dependent CH₄ emissions from wetlands between the two sites, which is deduced by analyzing their seasonal cycles with a 1-box model. Short-term CH₄ variations are observed throughout the year, showing an enhancement especially in summer. The relationships between the concentration and isotopic ratios for the short-term CH₄ variations yield the respective source signatures of δ¹³C and δD to be −63.4±2.8 and −316±24‰ for summer (May–October), and −47.7±4.5 and −244±52‰ for winter (November–April). These values suggest that the summertime and wintertime short-term CH₄ variations are produced by CH₄ emitted from wetlands and fossil fuel, respectively. The results simulated using an atmospheric chemistry transport model (ACTM) reproduce well the CH₄ variations observed at Ny-Ålesund, but much exceed the summertime CH₄
concentrations at Churchill. Tagged tracer experiments by the ACTM indicate that such high values are due to CH\textsubscript{4} emissions from the boreal zone in North America, suggesting that wetland fluxes calculated by a process-based ecosystem model (VISIT) to incorporate into the ACTM are overestimated for HBL.
Abstract:

The Arctic region is rapidly gaining interest and support for scientific studies to help understand and characterize the processes, sources, and chemical composition of the Arctic environment. In order to understand the Arctic climate system and the changes that are occurring, it is imperative to know the behavior and impact of atmospheric constituents. Surface level ozone in the Arctic is variable in both time and space and plays an essential role on the oxidation capacity of the atmosphere. NOAA Global Monitoring and Physical Sciences Divisions maintain continuous measurements and long-term records of ground-level ozone from Barrow, Alaska (since 1973), Summit, Greenland (since 2000), and Tiksi, Russia (since 2009). Measurements are quality checked and investigated with regards to wind conditions and aerosol loading. These quality controlled data are used to develop seasonal climatologies, understand diurnal variation, and analyze differences in stations specifics by addressing spatial variability in the Arctic. Once typical ozone behavior is characterized, anomalies in the record can be defined and investigated. Increased ozone events associated with transported pollution and photochemical production of ozone, and ozone depletion episodes related to sea-ice halogen release and chemical destruction of ozone are the primary processes which lead to deviations from expected ground-level ozone conditions. The measurements taken from Barrow (Alaska), Summit (Greenland), and Tiksi (Russia) are critical observations of ground-level ozone to provide fundamental understanding of the behavior and trends of ground-level ozone in the Arctic. The surface ozone and wind data are compared against Community Earth System Model (CESM1) CAM4-chem to assess model's ability to simulate surface ozone in the Arctic. An improvement agreement between observations and the chemistry-climate model hindcast is found when the model is forced with the reanalysis winds. Results are assessed by season, wind direction and impact of the local pollution.
Influence of anthropogenic emissions on the mass concentration and composition of organic PM1 in the Amazonian wet season (GoAmazon2014/5).

Early Career Scientist

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Abstract:

As part of GoAmazon2014/5, a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) was deployed to characterize the composition of fine-mode particulate matter (PM) and provide insights into the production of organic PM in the central Amazon basin, Brazil. Through a combination of meteorology, emissions, and chemistry, the T3 research site (located 70 km downwind of Manaus) was affected by biogenic emissions from the tropical rainforest that were periodically mixed with urban outflow from the Manaus metropolitan area as well as with biomass burning plumes. Results from the T3 site are presented in the context of measurements at T0a (ATTO) and T2, representing predominantly clean and polluted conditions, respectively. At T3, in the wet season (1/Feb 2014 - 31/Mar 2014) the non-refractory PM1 mass concentration had values on order of 1 to 2 μg m⁻³, and the organic component was dominant, contributing 79% by mass.

The analysis of the results aims at delineating the anthropogenic impact on the measurements, especially focusing on the effect of NOₓ emissions on the formation of organic PM. Positive matrix factorization (PMF) analysis is applied to the time series of
mass spectra of the organic component of PM$_1$. The resulting factors and their loadings provide information on the relative and time-varying contributions of different sources and processes of organic PM. The time trend of the different statistical factors is investigated against co-located measurements, and compared between background and polluted conditions. Results suggest that polluted conditions are associated with higher organic mass concentrations, with some PMF factors being favored under those conditions while others are inhibited. This analysis and results represent a step toward the goal of improving the understanding of anthropogenic influences on the mass concentrations and composition of PM$_1$ in Amazonia.
Spatial and temporal variability of in-situ and column integrated trace gases during DISCOVER-AQ.

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Abstract:
We quantify spatial and temporal variability of both in-situ mixing ratios and column integrated O$_3$, NO$_2$, CO, SO$_2$, and HCHO during the four deployments of the NASA Earth Venture mission DISCOVER-AQ (Deriving Information on Surface conditions from Column and Vertically Resolved Observations Relevant to Air Quality). These deployments consist of month-long campaigns over Baltimore, MD (Jul 2011), the San Joaquin Valley, CA (Jan-Feb 2013), Houston, TX (Sep 2013), and Denver, CO (Jul-Aug 2014). Using structure function analyses, we compare simulated variability of output from the regional chemical models WRF/Chem and CMAQ with variability observed during the campaigns. The Tropospheric Emissions: Monitoring of Pollution (TEMPO) instrument will be the first NASA mission to make atmospheric composition observations from geostationary orbit and partially fulfills the goals of the Geostationary Coastal and Air Pollution Events (GEO-CAPE) mission. Our analysis quantifies how often the atmosphere exhibits observationally relevant gradients in several key trace gas species, over what length scales they occur, and over what time periods. We compare both observed and simulated variability to the precision requirements defined by the science traceability matrices of these space-borne missions to explore how they relate to the science questions they are tasked to address.
Deployment of both dense networks and small numbers of low-cost and minimal infrastructure air quality sensors. A study in variable sensing scales.

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Abstract:
A major component of targeted air quality and associated measurement studies (e.g. for urban or industrial source apportionment, biomass burning plume extents or sea-breeze recirculation) is the better characterisation of horizontal scales of variability and relating these outputs to model and/or satellite products. There is particular interest in understanding how high spatio-temporal density in-situ data from emerging network systems can be reconciled with sub-satellite or model pixel scales (especially in high gradient areas and at pixel edges).

Data collected from high-density sensor networks during the NASA DISCOVER-AQ (Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality) campaign-based study are presented here alongside data from a dual sensor deployment in Malaysian Borneo with the aim of comparing site specific methodologies and outcomes.

As part of DISCOVER-AQ, autonomous, low-cost, high-density networks of air quality sensors were deployed. The 2 main deployments were 10 sensors for 3 months in Houston and 15 sensors for 5 weeks in Denver/Boulder (CO, NO, NO₂, SO₂, O₃, temperature and %RH at 1 minute resolution). These networks were designed to capture horizontal variability at very high spatial and temporal resolution. Vertical distribution at one site was also investigated in Colorado with sensors deployed on the 300m Atmospheric Observatory tower facility.
As part of an ongoing University Malaya study on low cost sensing technologies in logistically difficult environments, 2 sensors have been deployed for approximately 6 months at the Danum Valley research centre in Malaysian Borneo (relatively undisturbed dipterocarp forest). The sensors are located on the 75m station tower looking at potential trans-canopy concentration gradients (CO, NO, NO$_2$, SO$_2$, O$_3$, temperature and %RH at 1 minute resolution).

This work describes initial outcomes and selected data from these deployments and outlines ongoing studies (including current instrumental limitations) based on data collected.

Early Career Scientist

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Abstract:

Local variations in volatile organic compound (VOCs) and trace oxidizer concentrations can significantly impact haze and aerosol formation, albedo, and radiative forcing on a regional level. The use of unmanned aerial vehicles (UAVs) can be employed to sample trace gases in previously underrepresented regions of the troposphere. In order to put instrumentation on these UAVs, the use of lightweight, miniature sensors is critical to gather the necessary data yet meet weight requirements. This work will present the development of such a portable system with low-power requirement to track trace tropospheric gases. The sensors are first been evaluated under laboratory conditions to calibrate their response to target gases. We expect to present the first datasets for this system, which will be gathered during July 2016 in flights over Oklahoma that will quantify profiles of several gases (e.g., O$_3$ and NO$_2$), together with humidity, temperature, and pressure. An altimeter and real time clock provides correlation of the measurements to global positioning systems (GPS) alongside the sensing platform. Contour maps with the information gathered will be presented to display variations in each species over the area sampled. The presentation will provide the evolution of each species, which can be related to potential emission sources and effective exposure at ground level. In summary, the development and utilization of portable, sensitive sensor devices provides new perspectives in atmospheric sensing and quantification of trace gases.
6.044 Air pollutants in the lower troposphere over two rural sites in the North China Plain: results from ground-based and unmanned aircraft observations.

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Abstract:

Air pollution problem in the North China Plain (NCP) has been one of hot environmental issues for many years. Severe haze occurs frequently under unfavorable meteorological conditions, endangering human health and traffic safety in the densely populated region. The local authorities have been struggling against the high PM$_{2.5}$ level. Meanwhile, the photochemical pollution has emerged in the warm seasons, as indicated by high O$_3$ levels in the surface air. Air quality in the countryside of the NCP region can be also poor due to emissions from agricultural activities and transport of air pollutants from urban and industrial areas. Previous atmospheric chemistry studies in the NCP region have paid more attention to urban than to rural areas. To better characterize the major air pollutants and understand chemical and physical processes related to photochemical pollution in the rural areas, we performed field campaigns at the rural sites Gucheng (39°08'N, 115°40'E, 15 m asl) and Raoyang (115°44'E, 38°14'N, 20 m asl) in summer.
2013 and 2014, respectively. We observed photochemical pollutants and related species in the surface layer, including $O_3$, PAN, NO$_x$, NO$_y$, VOCs, HCHO, CO, SO$_2$, NH$_3$, PM, etc. We also made profile measurements of $O_3$, black carbon, and aerosol number concentration using devices attached to an unmanned aerial vehicle and a tethered balloon. Here we present and discuss the observational results. The variations of the ground-level gases are interpreted in terms of meteorology. The measurements of $O_3$, NO$_x$ and NO$_y$ are used to estimate ozone production efficiency. The vertical profiles of $O_3$ and particulate matter are discussed in relation to boundary layer evolution. Furthermore, the impact of convection process on surface $O_3$ is quantified.
6.045 Eastern Mediterranean transport patterns and atmospheric conditions associated with high and low summer ozone levels in the lower troposphere and the boundary layer.

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Abstract:

Vertical summertime ozone profiles measured within the MOZAIC project (1994–2008) over the Eastern Mediterranean airports of Cairo and Tel-Aviv were analyzed. Summer profiles during days with very high or very low ozone mixing ratios in the lower troposphere (1.5-5 km) have been examined together with the average profiles of relative humidity, carbon monoxide, temperature, wind speed and the corresponding composite maps of geopotential heights at 850 hPa. Also, profiles corresponding, respectively, to the highest and the lowest ozone mixing ratios for the 0-1.5 km layer over Cairo are examined along with their corresponding composite maps of the following meteorological parameters (and/or their anomalies): geopotential height, vertical velocity, specific humidity, columnar precipitable water, air temperature and wind speed at 850 hPa as well as the corresponding backward trajectories.

The principal result of this study is that the lower-tropospheric ozone variability over the eastern Mediterranean is controlled mainly by the synoptic meteorological conditions. In particular, the highest ozone concentrations in the lower troposphere and the boundary layer are associated with large-scale subsidence of ozone-rich air masses from the upper troposphere under anticyclonic conditions while the lowest ozone concentrations are associated with low pressure conditions inducing uplifting of boundary-layer air, poor in ozone and rich in relative humidity, to the lower troposphere (Kalabokas et al, ACP, 2013). Also, during the highest ozone days at the 0-1.5 km layer over Cairo, very high ozone concentrations around 80 ppb are observed from the surface up to 4-5 km altitude associated with extended geographical areas of strong subsidence and dry conditions over the eastern Mediterranean as well as over eastern and northern Europe under northerly flow (Kalabokas et al, TellusB, 2015), which in addition to the photochemical ozone production under the favorable Mediterranean summer conditions leads to high
summer surface ozone background levels.
Due to remarkable economic growth over the last two decades, East Asia has become a region experiencing some of the poorest air quality in the world. In addition to local sources of pollution, the Korea peninsula is downwind of the largest emission sources in East Asia, complicating the understanding of air quality over Korea. Thus, knowing the factors controlling changes in air pollution across urban-rural and marine-continental interfaces, in addition to the contributions from local emissions and transboundary transport, is important for building effective management strategies and improving air quality in East Asia.

During 6 weeks between May and June 2016, the KORea and United States Air Quality field study (KORUS-AQ) took place and employed an integrated observing strategy including multiplatform observations (i.e. ground stations, aircraft, ships, and satellites) and chemical transport models. This mission aims to not only strengthen our knowledge of atmospheric chemistry but also prepare for the future launch of geostationary satellite instruments by both countries to provide real-time monitoring of air quality (e.g. O3, NO2, SO2, HCHO, AOD, etc.) and enabling better scientific understanding of the transboundary transport of air pollutants. GEMS (Geostationary Environmental Monitoring Spectrometer) is a satellite instrument planned for launch in 2019 by the Republic of Korea. The instrument will observe East Asia and the western Pacific region. GEMS will also make
critical contributions to the global air quality perspective, working in concert with
geostationary missions launched by the U.S. (TEMPO: Tropospheric Emissions: Monitoring
of Pollution) and Europe (Sentinel-4) and low-Earth orbit missions including the European
Sentinel-5 Precursor. A scientific overview and expected outcomes from the mission will
be discussed in terms of the potential for improving both air quality models and the
remote sensing of air quality.
Thunderstorms transport trace gases and aerosols from the boundary layer to the upper troposphere and lower stratosphere (UTLS). Along the way, many processes occur, including lightning and its production of nitrogen oxides (NO\textsubscript{x}), scavenging by precipitation, chemical reactions, and transport. Consequently, high-altitude plumes of photochemically-active chemistry that produce ozone are found in the UTLS region, where ozone acts as a greenhouse gas. To advance our understanding of how thunderstorms affect tropospheric composition, the Deep Convective Clouds and Chemistry (DC3) experiment was conducted over the central US in May and June 2012. Here, we highlight recent results from the DC3 campaign.

As boundary layer air is ingested into thunderstorms, we expect most aerosols and some trace gases to be removed by clouds and precipitation. DC3 observations show that >80% of hygroscopic aerosols but only 10% of dust particles are removed during transport to the storm anvil regions, suggesting deep convection may be an efficient transport mechanism for dust. Scavenging efficiencies of formaldehyde and peroxides, important ozone precursors, are 41-58% (CH\textsubscript{2}O), >77% (H\textsubscript{2}O\textsubscript{2}) and 12-84% (CH\textsubscript{3}OOH). Lightning plays a very important role in atmospheric chemistry through its production of
NO\textsubscript{X}, which has been estimated for Colorado and Oklahoma DC3 storms to be 142-291 moles NO\textsubscript{X} per flash. We are beginning to connect these NO\textsubscript{X} production rates to lightning flash size. Estimates of ozone production in convective outflows with and without wildfire smoke underscore that more lightning-produced NO\textsubscript{X} creates more ozone, with 10-15 ppbv ozone produced during the day after convection. Although we expect ozone to be formed in the UT convective outflow plumes, a surprising result encountered during DC3 was the cloud-scale stratosphere-troposphere exchange caused by the thunderstorm dynamics. Measurements found stratospheric ozone alongside storm anvils, even wrapping around the anvil in one case, bringing high ozone concentrations into the UT.
In-service Aircraft for Global Monitoring: 20 Years of MOZAIC-IAGOS measurements.

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Abstract:

Climate change, air quality, and the oxidizing capacity of the atmosphere are major issues that require detailed, long-term observations of ozone and other atmospheric chemical compounds on a global scale. For already 22 years MOZAIC and its successor IAGOS have successfully harnessed the potential of in-service aircraft to respond to these needs. IAGOS is now a European Research Infrastructure established in 2014 (http://www.iagos.org) from two previous research projects, MOZAIC and CARIBIC, with the goal of establishing and operating a sustainable observing system for monitoring of atmospheric trace gases, aerosols and cloud particles from commercial aircraft at a global scale.

The presentation will give the ultimate goals of IAGOS, the current status of the technical implementation, and the planned developments. A strong focus will be made on the ozone, CO and water vapor measurements particularly in the upper troposphere and lowermost stratosphere (UTLS) at mid and high latitudes, along with the procedures applied to ensure high quality standards and consistency of the entire data set. The data quality control procedures have remained unchanged. Using overlapping years of MOZAIC/IAGOS, it has been shown that IAGOS can be considered as the continuation of MOZAIC with the same data quality.

A selection of scientific results from the past measurements will be presented in order to highlight the value of 20 years of regular airborne data from commercial aircraft for a better understanding of atmospheric composition along with its interannual variability and trends. IAGOS data are also widely used by the Copernicus Atmosphere Monitoring Service (CAMS) to evaluate forecast model runs in near real time as well as reanalysis. Examples of such process-oriented validation will be given. A collection of recent scientific achievements of the MOZAIC-IAGOS Programme was published as Special Issue of Tellus B; see http://www.tellusb.net/index.php/tellusb.

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Abstract:

SHADOZ (Southern Hemisphere Additional Ozonesondes) has collected more than 6000 profile sets from ozonesondes and radiosondes in the tropics and subtropics since 1998. Measurements originate at 14 long-term stations; map of the stations and data are archived at. Through affiliation with the Network for Detection of Atmospheric Composition Change (NDACC; www.ndsc.ncep.noaa.gov) and posting of profiles to the NASA Aura Validation Data Center and WMO’s World Ozone and UV Data Centre (woudc.org), SHADOZ data are distributed across the satellite, monitoring and modeling communities. We review major activities of SHADOZ since the last IGAC, including re-activation of five SHADOZ stations: Ascension, Fiji, Irene, San Cristobal, Natal. Examples of newer data from those sites will be shown. The most significant SHADOZ activity we have recently carried out is the first major reprocessing of the 18-year ozonesonde dataset to account for changes in radiosonde and ozonesonde instrumentation and biases among stations. The reprocessing has been done following guidelines of WMO and SI2N (SPARC/Intl Ozone Commission/IGACO and NDACC). Impacts of re-processing on agreement between satellite profiles and total ozone and the corresponding sonde measurements will be displayed along with re-examination of station biases in the stratospheric portion of the ozonesondes.
Sixteen year CO emission estimates constrained with MOPITT CO measurements.

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Abstract:
Long-term measurements from satellite and surface stations have demonstrated a decreasing trend of tropospheric CO in Northern Hemisphere in the past decade, and the major reason is still unclear. Using remote sensing CO measurements from Measurement of Pollution in the Troposphere (MOPITT), and in-situ methyl chloroform (MCF) measurements from World Data Centre for Greenhouse Gases (WDCGG), we investigate the major reason for the decreasing tropospheric CO in the period of 2001-2015. Our results demonstrate that the loss rate of MCF is almost invariable in the past 15 years, suggesting unchanging OH abundance (CO sink). Using adjoint of GEOS-Chem model, we constrain the global CO emission variation. Our results demonstrate significant decreasing trend of anthropogenic emissions from United States, Europe and China, and a decreasing trend of biomass burning emissions from boreal North America and boreal Asia. The a posteriori surface CO magnitude and long-term trend are evaluated with independent in-situ CO observations from WDCGG, which show significant improvement relative to the a priori simulation. Our results confirm the decreasing trend of tropospheric CO in Northern Hemisphere, and indicate that decreasing CO emissions from anthropogenic and biomass burning sources are the major reason.

Early Career Scientist

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Abstract:

Ammonia is the third most abundant nitrogen containing species in the atmosphere. Its reaction with sulfuric acid (H$_2$SO$_4$) forms ammonium sulfate (NH$_4$)$_2$SO$_4$ or ammonium bisulfate (NH$_4$)HSO$_4$ aerosols, two important types of particulate matter (PM$_{2.5}$) which can have harmful effects on human health and the environment. Measurements of atmospheric ammonia are challenging because it is a trace species present with widely varying concentration, and because it is a sticky gas, such that there are significant inlet challenges for sampling techniques that involve direct contact between the sample gas and inlet or flow cell materials. An open-path absorption spectrometer can eliminate sampling issues since the ambient air flows freely across the probe laser beam. Removing the inlet can also be enabling for compact and low power designs, for example for use on unmanned aerial systems (UAS). This contribution presents research on two open-path ammonia laser sensors both of which target the strongest absorption features of ammonia in the 9-10 μm region using mid-infrared quantum cascade lasers (QCLs). First, we detail an open-path cavity ring-down spectroscopy (CRDS) instrument. This sensor shows a detection limit of 0.8 ppb (in 2 seconds) and good agreement with a commercial closed-path CRDS sensor in a side-by-side comparison at a feedlot. Second, we detail a follow-on instrument based on wavelength modulation spectroscopy (WMS) which seeks to achieve smaller mass and power. A multi-pass cell (30 cm base path with 20 m path length) is used to enhance sensitivity. All electronics are miniaturized such that the whole sensor size is compact (~3 kg) for UAS deployment. The contribution will discuss upcoming UAS measurements with the WMS sensor along with a CRDS methane sensor for agricultural emissions. Methane will be used as a conservative tracer and spatial variations in the ammonia-methane ratios will be used to study bi-directional ammonia fluxes.
Aromatic VOC effects on OH and Ozone at global scale.

Early Career Scientist

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Abstract:

Volatile Organic Compounds (VOC) are partially responsible of the formation and removal of atmospheric key species as ozone and hydroxyl radical (OH). Among VOCs, aromatics compounds concentrations make up for a large VOC fraction in urban and peri-urban areas (up to 60%). Aromatic sources are mainly related to fossil fuel use and biomass burning, and they are removed primarily by reaction with OH and secondarily by dry deposition. The lifetimes range from hours to days, hence they may be transported to areas far from sources and influence air quality there.

The effect that aromatic have on ozone and OH formation/destruction is poorly understood.

This work assesses the atmospheric impacts of the most commonly emitted aromatic VOCs on OH and ozone at global scale, with the help of an atmospheric chemistry general circulation model. Specifically, the percentage of ozone produced/removed due to aromatic oxidation is of great interest in large urban areas and can be helpful for developing air pollution control strategies. Furthermore, We disentangle in which areas OH formation is enhanced or depleted and unravel the mechanism controlling it.

Further targets are the quantification of the fraction of aromatics that leads to CO and second organic aerosol formation. In order to investigate the sensitivity of ozone and OH to aromatics, two simulations are performed with the atmospheric chemistry-climate model ECHAM/MESSy (EMAC). One that includes the presence of aromatics and one without them.
Abstract:

Lightning flashes heat the air to temperatures exceeding 20,000 K dissociating molecular oxygen and nitrogen, which then combine to create nitric oxide (NO), which quickly reacts with molecular oxygen to form nitrogen dioxide (NO\textsubscript{2}), a strong absorber in the visible that is measured from space by the Ozone Monitoring Instrument (OMI) aboard NASA's Aura satellite. Recently, we developed an algorithm to retrieve the lightning NO\textsubscript{x} (LNO\textsubscript{x}) signal from OMI and have applied it to estimate LNO\textsubscript{x} production efficiency (moles of NO\textsubscript{x} per flash) over the Gulf of Mexico for June-August 2007-2011 using detection-efficiency adjusted flash data from the ground-based World Wide Lightning Location Network (WWLLN) and Vaisala Global Lightning Data 360 (GLD360) networks. Currently, we are expanding the analysis regions to include central- and western-Africa, the western Pacific, and the Amazon Basin and the time periods to include September-November 2010-2012. Conceptually, the algorithm takes the total slant column NO\textsubscript{2} from OMI, removes the stratospheric contribution and tropospheric background (NO\textsubscript{2} from sources other than recent lightning) and then calculates the LNO\textsubscript{x} vertical column from the LNO\textsubscript{2} slant column using an appropriate air mass factor. In practice, the algorithm is applied to locations where the OMI cloud radiance fraction exceeds 90\% suggestive of active convection. In these regions, the background contribution to the column is believed to be small. In this presentation, we will show results for several of these regions with an emphasis on quantifying uncertainties in the estimates. These uncertainties include random and systematic errors in OMI slant columns, the stratospheric vertical column amount, WWLLN and GLD360 detection efficiencies, the OMI Optical Centroid Pressure below which OMI cannot see LNO\textsubscript{x}, the flash counting window, CRF threshold, and the approach used to estimate the tropospheric background.
6.062 Measurements of organic composition of aerosol and rainwater samples using offline aerosol mass spectrometry.

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Abstract:

A thorough understanding of the sources, evolution, and budgets of atmospheric organic aerosol requires widespread measurements of the amount and chemical composition of atmospheric organic carbon in the condensed phase (within particles and water droplets). Collecting such datasets requires substantial spatial and temporal (long term) coverage, which can be challenging when relying on online measurements by state-of-the-art research-grade instrumentation (such as those used in atmospheric chemistry field studies). Instead, samples are routinely collected using relatively low-cost techniques, such as aerosol filters, for offline analysis of their chemical composition. However, measurements made by online and offline instruments can be fundamentally different, leading to disparities between data from field studies and those from more routine monitoring. To better connect these two approaches, and take advantage of the benefits of each, we have developed a method to introduce collected samples into online aerosol instruments using nebulization. Because nebulizers typically require tens to hundreds of milliliters of solution, limiting this technique to large samples, we developed a new, ultrasonic micro-nebulizer that requires only small volumes (tens of microliters) of sample for chemical analysis. The nebulized (resuspended) sample is then sent into a high-resolution Aerosol Mass Spectrometer (AMS), a widely-used instrument that provides key information on the chemical composition of aerosol particulate matter (elemental ratios, carbon oxidation state, etc.), measurements that are not typically made for collected atmospheric samples. Here, we compare AMS data collected using standard on-line techniques with our offline analysis, demonstrating the utility of this new technique to aerosol filter samples. We then apply this approach to organic aerosol filter samples collected in remote regions, as well as rainwater samples from across the US. This data provides information on the sample composition and changes in key chemical characteristics across locations and seasons.
6.065 Measurements of nitrous acid (HONO) within the clean tropical marine boundary layer.

Early Career Scientist

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Abstract:

At ground level HONO is an important source of OH radicals - yet its sources and formation mechanism are poorly understood, in particular in the marine boundary layer environment which dominates the global surface. Measurements of HONO were conducted at ground level at the Cape Verde Atmospheric Observatory (CVAO), during November 2015. During this time, Cape Verde experiences frequent dust storms, originating from continental Africa. In addition, complementary gas-phase, particle phase and meteorological measurements were obtained. Overall low concentrations of HONO were observed (<6 ppt). However a distinct diurnal variation was observed, characterised by peak in concentration at midday and concentrations below the detection limit (<1 ppt) at night, and which varied with photolysis frequency (i.e. degree of haze). A similar diurnal trend was also observed for NO, whereas the NO\textsubscript{2} concentration was relatively constant. The potential for different HONO formation sources, including heterogeneous and photo-enhanced processes, at CVAO and hence in the wider tropical marine boundary layer, are explored.
Measuring Atmospheric CO2 with the NASA Orbiting Carbon Observatory-2 (OCO-2).

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Abstract:

Fossil fuel combustion, deforestation, and other human activities are adding almost 40 billion tons of carbon dioxide (CO2) to the atmosphere each year. This is enough CO2 to increase the atmospheric concentration of this gas by 1% per year. Interestingly, less than half of this CO2 stays airborne, on average. The rest is apparently being absorbed by natural processes at the surface, whose identity and location are poorly understood. Ground-based CO2 measurements accurately record the global atmospheric CO2 budget and its trends, but do not have the spatial resolution or coverage needed to identify the “sources” emitting CO2 into the atmosphere or the natural “sinks” absorbing this gas at the surface.

One way to improve the resolution and coverage of these measurements is to collect precise observations of CO2 from an orbiting satellite. The Orbiting Carbon Observatory-2 (OCO-2) is NASA’s first satellite designed to address this need. OCO-2 was successfully launched on July 2, 2014. By early September of 2014, its high resolution imaging grating spectrometers were recording almost a million soundings over Earth’s sunlit hemisphere each day. Observations recorded over the first 18 months of operation clearly show the most robust features of the atmospheric carbon cycle, including the intense northern hemisphere spring drawdown, as land plants rapidly absorb CO2 to form new leaves, stems, and roots. They also show enhanced CO2 over regions with intense fossil fuel combustion, such as the east coast of China and the U.S. As these measurements are analyzed by the carbon cycle science community, they are expected to reveal far more detail about the processes controlling the atmospheric CO2 buildup. This talk will describe the OCO-2 mission, summarize its measurement approach, and present results from its first 18 months in operation.
Abstract:

The return of sunlight to the polar regions is associated with the release of molecular halogens from the snowpack. The photolysis of these halogens produces halogen radicals, leading to depletion of boundary layer ozone to near-zero levels and altering oxidation of atmospheric pollutants, particularly gaseous elemental mercury. Despite measurements of ozone depletion events over several decades, our understanding of the vertical extent of this halogen chemistry remains limited. During the 2012 BRomine Ozone Mercury EXperiment (BROMEX), the Purdue Airborne Laboratory for Atmospheric Research (ALAR) conducted 8 flights to measure vertical profiles of BrO, ozone, and aerosol particle number concentrations. These flights showed ozone depleted regions and halogen enriched regions generally having similar vertical extents. BrO was observed both near the snowpack and aloft, sometimes independently. MODIS imagery of local sea
Ice dynamics is used to investigate the influence of enhanced vertical mixing associated with open sea ice leads on the vertical distribution of reactive halogens and ozone. The role of aerosol particles in sustaining halogen activation aloft will also be discussed.
6.074 Quantifying wildfire emissions and associated aerosols species using assimilation of satellite carbon monoxide retrievals.

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Abstract:

Intense and costly wildfires tend are predicted to increase in frequency under a warming climate. For example, the recent August 2015 Washington State fires were the largest in the state’s history. Also in September and October 2015 very intense fires over Indonesia produced some of the highest concentrations of carbon monoxide (CO) ever seen from satellite. Such large fires impact not only the local environment but also affect air quality far downwind through the long-range transport of pollutants. Global to continental scale coverage showing the evolution of CO resulting from fire emission is available from satellite observations. Carbon monoxide is the only atmospheric trace gas for which satellite multispectral retrievals have demonstrated reliable independent profile information close to the surface and also higher in the free troposphere. The unique CO profile product from Terra/MOPITT clearly distinguishes near-surface CO from the free troposphere CO. Also previous studies have suggested strong correlations between primary emissions of fire organic and black carbon aerosols and CO. We will present results from the Ensemble Adjustment Kalman Filter (DART) system that has been developed to assimilate MOPITT CO in the global-scale chemistry-climate model CAM-Chem. The ensemble technique allows inference on various fire model state variables such as CO emissions, and also aerosol species resulting from fires such as organic and black carbon. The benefit of MOPITT CO profile assimilation for estimating the CO emissions from the Washington and Indonesian fire cases will be discussed, along with the ability of the ensemble approach to infer information on the black and organic carbon aerosol distribution.

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Abstract:

Mineral dust aerosol particles impact the Earth’s radiative balance via direct scattering and absorption of light and by promoting ice cloud formation. Modelling studies suggest that mineral dust is a leading contributor to aerosol optical depth throughout much of the globe. Lab and field studies indicate that dust particles are efficient ice nuclei, and recent airborne measurements confirm the dominant role of mineral dust on cirrus cloud formation in the Northern Hemisphere. However, our ability to simulate the net climate impact of mineral dust is limited by a lack of validating measurements of dust abundance and mixing state above the Earth’s surface. We present a summary of mineral dust measurements in the free troposphere from more than 10 years of airborne sampling, highlighting vertical distributions, hemispheric dissimilarities, chemical transformation, and dust’s role in cirrus formation. Clear sky aerosol measurements show that mineral dust is ubiquitous in the background upper troposphere at levels from ~1 to 1000’s per liter and frequently accounts for 5-40% of particulate mass. Principal sources of upper tropospheric mineral dust include deep convection, strong biomass burning events, and long range isentropic transport. Over time mineral dust becomes coated with secondary sulfate, nitrate, and organic material that can deactivate ice nucleation sites on particle surfaces. This reduction in cirrus-forming potential depends on co-emissions, transport mechanisms, and particle age.
6.078 Convective Influence on Methanol (CH3OH) in the Tropical Upper Troposphere.

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Abstract:

Long-term measurements of methanol (CH$_3$OH) from the Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS) together with in-situ measurements from the DC3 (2012) and SEAC$^4$RS (2013) field campaigns conducted over North America are used to better understand its variability in the upper troposphere. The main objective of this study is to investigate the influence of surface biogenic emissions and convective transport on methanol distributions over different regions and season. Simulations of methanol from the Community Atmosphere Model with Chemistry (CAM-chem) global chemistry model are also used to evaluate the model with a focus on Asian vs. North American summer monsoons. The results show overall agreement in the shape of the vertical distributions of methanol from the measurements and the model, reflecting the transport of directly emitted methanol to the upper troposphere. Differences in the magnitudes of the model simulations and satellite measurements are due to an underestimate of emissions, as supported by model evaluation with the aircraft observations.
6.079 Quantifying information content and gaps in the AERONET network in South America.

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Abstract:

In previous work we have developed and applied statistical and variational approaches to characterize the representativity and specificity of air quality networks, as well as the changes in information content over time. These methods are derived from information theory and used in the context of optimal network design. Here we apply these methods to better characterize the atmospheric optical depth (AOD) information collected from an increasing number of sun photometers under the Aerosol Robotic Network (AERONET) in South America. Thus in addition to assessing how the network’s information content has grown over time, and quantifying the representativity and specificity of the stations, we gain insight in terms of the suitability of the network stations for data assimilation exercises. Preliminary results will be presented.
Utilizing model sensitivity analysis to set research priorities and constrain the global acetone budget.

Early Career Scientist

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Abstract:

Acetone is one of the most abundant carbonyl compounds in the atmosphere, and a major source of HOX radicals in the upper troposphere. Thus, understanding the global budget of acetone is essential to understanding global oxidation capacity. Significant uncertainties remain regarding the flux of acetone out of and into the biosphere. Crucially unconstrained processes include dry deposition, fluxes of acetone into and out of the ocean, direct emissions of acetone from the terrestrial biosphere, direct emissions of secondary sources of acetone such as the oxidation of monoterpenes from the terrestrial biosphere, and loss of acetone due to photolysis.

We have performed an elementary effects sensitivity analysis of the GEOS-Chem global 3-D CTM (version 10-01, www.geos-chem.org) for the global atmospheric distribution of acetone using the Morris method. This method provides a ranking of both the comparative direct importance, as well as non-linear effects and interactions of the tested input factor uncertainties, at a relatively low computational cost. The sensitivity analysis was bounded using literature minima and maxima for six sources of uncertainty related to specific sources and sinks. The uncertainties with the largest impact on acetone concentration are the uncertainties in direct acetone emissions from the terrestrial biosphere, the uncertainties in modeled j-values used to represent acetone photolysis, and the uncertainties in the concentration of acetone in the ocean mixed layer. Future work to improve the global acetone budget should therefore be primarily focused on improving the representation of these three processes within global CTMs.
6.085 Distributions of BrO and IO in the tropical marine boundary layer and free troposphere.

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Abstract:

Halogens affect atmospheric chemistry by destroying ozone, changing the oxidative balance, processing mercury, and by particle growth. The halogen radicals bromine and iodine monoxide (BrO and IO) were measured by the University of Colorado Airborne Multi-AXis DOAS instrument (CU AMAX-DOAS) over the Western (CONTRAST project) and Eastern (TORERO project) tropical Pacific Ocean aboard the NSF/NCAR GV aircraft. Observations of up to 3.0 pptv BrO and 0.1 to 0.25 pptv IO over the Eastern Pacific Ocean (Dix et al., 2013; Volkamer et al., 2015; Wang et al., 2015) pose new constraints to develop halogen chemistry in state-of-the-art models, which tend to underestimate the total BrO column, and over predict BrO in the lower troposphere.

CU AMAX-DOAS is optimized to (1) locate BrO, IO, and the OVOC glyoxal in the troposphere, (2) decouple stratospheric absorbers, (3) maximize sensitivity at instrument altitude, and (4) enable observations over a wide range of SZA. In the past we have used Optimal Estimation inversion to retrieve vertical BrO and IO profiles during aircraft ascents and descents that were successfully evaluated by comparing e.g. DOAS NO2 and H2O with atmospheric models and in-situ observations (Volkamer et al., 2015). We have also developed a method that parametrizes radiative transfer for the fast conversion of slant column densities into volume mixing ratios along the flight track. The method is based on comparison of O2-O2 collision complexes at tropospheric altitudes. Here we present a summary of BrO and IO distributions based on AMAX-DOAS observations over the Western and Eastern tropical Pacific, describe our methods, discuss spatial distributions and atmospheric implications. Further we introduce the concept of mountain top measurements in the tropical free troposphere, to generate long-term time series for model evaluation of a critical and data poor compartment of the atmosphere.
6.088 PERSEUS: A SAMPLE PRECONCENTRATION AND GC/MS DETECTOR SYSTEM FOR ANALYSIS OF FLASK AIR SAMPLES FOR ATMOSPHERIC TRACE HALOCARBONS, HYDROCARBONS AND SULFUR-CONTAINING COMPOUNDS.

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Abstract:
A newly developed analyte preconcentration system for GC/MS expands the volatility range of the atmospheric halocarbons, hydrocarbons and sulfur-containing compounds measured in the long-term monitoring program of the Global Monitoring Division. Ethane (C2H6), tetrafluoromethane (CF4), nitrogen trifluoride (NF3), hexafluoroethane (C2F6), trifluoromethane (CHF3), sulfuryl fluoride (SO2F2) and toluene (C7H8) are among the new additions to the 50+ greenhouse gases, ozone depleting substances and/or species of air quality involvement that are routinely measured in flask samples collected semi-daily at towers and semi-monthly in tropospheric altitude profiles with small aircraft across North America. Weekly flask samples collected at more remote ‘clean air’ sites globally serve as a background against which North American enhancements may be calculated. We describe here the technological development of this new analytical system, discuss its expanded capabilities and present example case studies from the first two years of atmospheric records.
6.093 Impact of the summer circulation regimes on the vertical tropospheric distribution of pollutants and aerosol processes over the Western Mediterranean area.

Early Career Scientist

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Abstract:
This work investigates the effects of different meteorological conditions on the vertical distribution of aerosol and chemicals over the Po Valley air pollution hot spot. This region is strongly affected by various natural and anthropogenic sources and its morphology, being a river basin surrounded by mountains, leads to frequent stagnation and pollutants accumulation. The unique and comprehensive observations database built by the ChArMEx (Chemistry-Aerosol MEditerranean Experiment), PEGASOS (Pan-European Gas-AeroSOls Climate Interaction Study) and SuperSito campaigns, simultaneously operating in the Mediterranean area during summer 2012, was exploited to carry out an aerosol discrimination based on LIDAR profiles and aerosol and chemistry in-situ observations. A broader view on the processes was provided by model analysis from the Lagrangian transport system FLEXPART and WRF-Chem, the Weather Research and Forecasting (WRF) model coupled with Chemistry. Results showed that particles were mostly individuated below 2000m, with a prevalence of local anthropogenic aerosol (> 50% of observations) and a bi-modal particle diameter (Dp) size distribution (Dp<1 um and 2um80%), both processes affecting the local Particulate Matter concentration. The combination of well-developed PBL and favourable winds allowed an event of pollution outflow, with air parcels overcoming the orography barriers and being transported toward
the Western Mediterranean sea for several hundred km. The plume, traced with CO concentrations, extended vertically up to 1000m and carried an average contribution of 50 ppbv over the background values.
6.095 Atmospheric CH4 mixing ratios and its correlation with CO and a surrogate of Non-Methane Hydrocarbons in Mexico City.

Early Career Scientist

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Abstract:
Methane (CH$_4$) is one of the greenhouse gas (GHG) emitted by both natural and anthropogenic sources. It has been mentioned that, megacities could account for around 7-15% of their anthropogenic CH$_4$ emissions and that it is possible that these came from fugitive emissions from the natural-gas supply lines. The Mexico City Metropolitan Area (MCMA) is one of the world’s largest and most polluted urban areas; although natural gas is not the most used fuel for domestic, industrial heating or transport in MCMA. A recent GHC emission inventory for MCMA suggests that vehicular emissions contribute with approximately 37% of CH$_4$, followed by landfills and dump garbage areas (30%) and construction and manufacturing (27%). An intensive monitoring campaign of CH$_4$, Non-methane hydrocarbons (NMHC) and carbon monoxide (CO) was performed between February and March 2015 in southwest MCMA in order to understand the role of vehicular emissions on the atmospheric methane levels. Methane concentrations showed sometimes a diurnal pattern similar to those of CO and to NMHC but the similarity between the peaks was lost (CH$_4$ vs CO, $R^2 = 0.27$; CH$_4$ vs NMHC, $R^2 = 0.28$). However, NMHC correlated well with CO ($R^2 = 0.75$). The intercepts of the CH$_4$-CO correlation resulted in [CH$_4$] $\sim$ 1.8 ppm and of the CO-NMHC correlation in [CO] $\sim$ 0.080 ppb. A comparison with historical CH$_4$ measurements in MCMA showed that the mean methane concentration observed in March, 1993 was around 3.1 ppm while during the 2015 campaign it was 1.93 ppb. The lowest average of CH$_4$ levels in 1993 was around 1.88 ppm. It seems that vehicular emissions are not significant contributors to atmospheric CH$_4$ and that the historic methane concentration has not change significantly in 25 years.
Assessing the impact of the anthropocene on atmospheric composition using remote sensing from aircraft and space based instrumentation..

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Abstract:

Since the industrial revolution the population of the earth has grown to ~7.5 Billion, urbanized and at the same time its standard of living has increased dramatically. This has been made possible through the exploitation of fossil fuels as an energy source. Since the 1950s and in spite of legislation successes in North America and Europe, there has been globally an acceleration of the release of both short lived climate pollutants and long lived greenhouse gases. Air quality, stratospheric ozone and climate change are all being influenced by anthropogenic activity and the earth has entered a new geological epoch the Anthropocene. The SCIAMACHY (Scanning Imaging Absorption spectrometer for Atmospheric CHartographY) project, which began in the 1980s, aimed to yield the amounts and distribution of atmospheric constituents: trace gases, aerosols and clouds from space based passive remote sensing in the solar spectral range. As a result of this initiative the following instruments have been developed and launched on satellite platforms into sun synchronous low earth orbit: GOME (Global Ozone Monitoring Experiment - ESA ERS-2 1995-2011), SCIAMACHY (ESA Envisat 2002 to 2012) GOME_2 (EUMTSAT Metop A 2006 to present, Metop B 2012 to present). In addition the spin off OMI (Ozone Monitoring Instrument - NASA Aura 2004 to present) was developed by NSO. Appropriate mathematical inversion of the measurements of these instruments yields information about the total stratospheric and tropospheric column amounts and distributions of the following gases, which are either short lived climate pollutants or greenhouse gases: O3, NO2, H2O, HCHO, CHO.CHO, BrO, IO, CO, CH4 and CO2. This presentation will provide an update on the analysis at the University of Bremen of the time series provided by these instruments and new measurements from related
instruments developed for aircraft.
Abstract:

Ozone concentrations at the earth’s surface are influenced by both meteorological and chemical processes and are a function of advection, vertical mixing, deposition, chemical production and loss. Understanding the relative importance of these mechanisms controlling surface ozone concentrations is an essential component for designing effective control strategies. However, determining the magnitude of these processes contributing to the total ozone budget generally comes with high uncertainties resulting from the diversity of measurements required. Here we combine ozone vertical profiles measured in summer 2014 in the Colorado Front Range during the DISCOVER-AQ and FRAPPE campaigns with surface measurements of ozone and planetary boundary layer height (PBLH). Vertical profiles obtained 3 times a day at 6 different sites allow us to analyze the diurnal evolution, day-to-day and site-to-site variability of vertical mixing’s influence on observed ozone concentrations in Colorado.

Above boundary layer concentrations can lead to both diluting and polluting effects of surface ozone depending on the sign of the concentration jump at the top of the PBL. Overall vertical mixing was found to pollute the surface in the morning and dilute in the afternoon. The time period in between can be characterized as a transition period that is dependent on the day-to-day variability of ozone concentrations in the residual layer and the free troposphere.

We also investigate vertical mixing of ozone in a WRF-CHEM-3D simulation over Colorado compared to the observations. We use additionally WRF-CHEM run as a single column model (WRF-CHEM-SCM) to interrogate vertical mixing in WRF-CHEM for idealized cases to understand the influence of boundary layer height, vertical resolution and emissions on predicted surface ozone concentrations. The work therefore lends to both regional and global importance, as we demonstrate how vertical mixing in WRF-CHEM can be studied.
in an idealized framework toward an understanding that can be translated across regions.
Polycyclic aromatic hydrocarbons (PAHs) are regarded as persistent organic compounds which is emitted by incomplete combustion process from fossil fuel, vehicles, and biomass burning. PAHs are highly concerned because of carcinogenic and mutagenic properties. Considering that the rapid population growth and energy demand, economical development, East Asia is one of the large PAHs emitted region. This study analyzed temporal variations of particulate PAH, organic carbon (OC), elementary carbon (EC) concentrations at Noto, facing the Sea of Japan. Particulate phase PAH, OC, and EC concentrations are clear seasonal variation with high in winter and low in summer. The PAH concentrations in winter at Noto are increased until the year of 2008, after then, decreased. The concentrations of OC and EC are increased before 2006 and then decreased until 2008, after then increased. After the year of 2011, the concentrations are decreased. It is found that PAH concentrations are increased with increasing EC as well as OC concentrations. The relation was different among seasons. In winter, PAH concentrations against EC or OC are larger than those in other seasons. The ratio of OC/PAHs and EC/PAHs are high in the period from 2004 to 2006 and after then rapidly decreased. These suggest that the PAH emissions are changed reflecting the emission amount as well as source contribution. The effect of changing of source contribution for PAHs, OC, and EC and temporal variations of these concentrations will be also discussed using the chemical transport model, Regional Air Quality Model (RAQM-POPs).

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Abstract:

Urban areas are responsible for a major component of the anthropogenic greenhouse gas (GHG) emissions. Quantification of urban GHG fluxes is important for establishing scientifically sound and cost-effective policies for mitigating GHG emissions. Discrepancies between observations and model simulations of GHGs suggest uncharacterized sources in urban environments. In this study, we quantified fluxes of carbon dioxide (CO$_2$) and methane (CH$_4$) from the Baltimore-Washington and Indianapolis areas based on the mass balance approach using observations from two aircraft flown in winter 2015 and 2016. Using the HYSPLIT back trajectory model, point sources of CO$_2$ and CH$_4$ were identified. The emissions from these point sources were quantified based on the aircraft observations and compared to the emission inventory data. In limited canister samples collected during the flights, the ethane-to-methane ratios indicate that both natural gas leaks and the upwind oil and natural gas operations are responsible for the observed enhancement in CH$_4$ concentration downwind of the urban area. The CO$_2$ data collected during the campaign are compared to OCO-2 satellite observations.
6.106 Quantifying isotopic signatures of atmospheric NOx emissions.

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Abstract:

Atmospheric nitrogen oxides (NO\textsubscript{x} = NO + NO\textsubscript{2}) play key roles in atmospheric chemistry, air quality, and radiative forcing, and contribute to nitric acid deposition. Given the variety of sources of NO\textsubscript{x} (both natural and anthropogenic), their variability in space and time, and the relatively short lifetime of NO\textsubscript{x}, it is difficult to directly link variability in NO\textsubscript{x} concentrations or nitric acid deposition with emissions sources. NO\textsubscript{x} isotopic signatures offer a potentially valuable tool to trace source impacts on atmospheric chemistry and regional acid deposition. However, previous work on NO\textsubscript{x} isotopic signatures suggests large ranges in values, even from the same emission source, as well as overlapping ranges amongst different sources. These prior measurements have utilized a variety of methods for collection of NO\textsubscript{x}, and recent tests reveal inconsistencies in efficiency of collection, as well as issues related to changes in conditions such as humidity, temperature, and NO\textsubscript{x} fluxes. Our recently developed method accurately quantifies the nitrogen isotopic composition (δ\textsuperscript{15}N) of NO\textsubscript{x} (NO\textsubscript{x} = NO + NO\textsubscript{2}) after capturing the NO\textsubscript{x} in solution as nitrate. The method has been thoroughly laboratory and field tested, and efficiently collects NO and NO\textsubscript{2} under a variety of conditions. This new method is used to quantify the isotopic composition of NO\textsubscript{x} associated with different emission sources, including vehicles, microbial processes in agricultural soils, and biomass burning. The NO\textsubscript{x} collection system is optimized to allow for short collections in high concentration plumes, as well as under sporadic pulse-oriented fluxes such those associated with soil emissions. Using a consistent method, we test whether it is possible to distinguish the isotopic ranges associated with different emission sources, and compare and contrast laboratory-based and field-based collections.
6.107 Tracking pollutants from space: 10 years of IASI satellite observation.

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Abstract:

The IASI instrument is a Fourier Transform Spectrometer sounding the atmosphere using the thermal infrared spectral range from a polar orbiting satellite. It was designed by CNES (Centre National d’Etudes Spatiales) and launched by Eumetsat on the MetOp series of satellites. The mission has been providing global observations of the air composition with an excellent spatial resolution, twice a day since 2007 (MetOp-A) and 4 times per day since 2012 (Metop-A and MetOp-B).

From the atmospheric spectra recorded by the instrument, concentrations of several trace gases can be monitored, enhanced levels of pollution can be detected, and particle types can be determined to some extent. This paper recalls the historical context for the IASI remote sensor, reviews its capability to observe some key species for global and regional pollution monitoring, and reports on information services that benefit from the mission. On the longer term the continuity of the program is ensured with the IASI-NG mission that will extend the IASI observation for 15-20 more years.
The increase of greenhouse gases (GHGs) concentrations in the atmosphere has been recognised as a major driver of climate change (IPCC, 2013). In response to this, the International community agreed in cutting anthropogenic emissions of GHG through the Kyoto Protocol, an international treaty linked to the United Nations Framework Convention on Climate Change (UNFCCC), which commits its Parties by setting internationally binding emission reduction targets. The reliability of the emissions data reported by the Parties to UNFCCC, is crucial in assessing the Parties compliance to the Kyoto Protocol and therefore in determining the Protocol’s effectiveness. Following the IPCC guidelines, the UNFCCC requires only bottom-up reporting, which relies on declared activities from industrial processes and product use and on emission factors for the release into the atmosphere. The availability of independent data, such as measurements of GHG atmospheric concentrations to be used for emission modelling, the so-called top-down approach, has been recognised as an important verification tool. Among the Kyoto gases, HFCs are considered particularly suitable for inverse modelling verification, being solely anthropogenic, long-lived and characterised by a well-known removal mechanism and such verification would be particularly important being their inventories affected by considerable uncertainties.

In this work we estimate, using atmospheric data from four European sites combined with a Bayesian inversion method, the emissions of nine individual HFCs from Europe and from twelve group of countries, comparing our results with the annual reports that the European countries submit to the UNFCCC and with the Emissions Database for Global Atmospheric Research. The observed discrepancies are then discussed. The reference period is from 2002 to 2014. The estimates provided by this analysis are relevant for constraining the atmospheric budget of these gases on a regional scale, also improving the accuracy of their emissions quantification on a global scale.
6.110 Influence of winds on retrievals of space based measurements of NO2.

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Abstract:

Satellite remote sensing of provides beautiful maps of the tropospheric column NO2 and their trends. Interpretation of the observed spectrum requires an initial guess, the a priori, about the vertical distribution of NO2 in the observed column. Here we will review why high spatial resolution (~4-12km) a priori is essential to an accurate retrieval and how wind speed affects the lifetime of NO2. We will then show that there are substantial improvements in the accuracy of a retrieval--at high spatial resolution--if daily instead of monthly average a priori fields are used. Consequences of these improvements for estimates of emissions will be described. We find emission estimates change by as much as 10-30% using a priors that correctly capture the effects of the boundary layer winds.
6.114 Transport and transformation of trace species in a deep convective cloud - results of the ACRIDICON tracer experiment.

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Abstract:

We report on a perfluorocarbon tracer experiment performed during the HALO (German GV research aircraft) campaign ACRIDICON in Manaus, Amazonas, Brazil, on 25 Sept. 2014 to study air mass transport and transformation of trace species in a deep tropical convective cloud. The tracer experiment included three steps: tagging of the pollution plume of the city of Manaus with the artificial gas tracer PMCH (C$_7$F$_{14}$), sampling of the tracer in the boundary layer downstream of the release site, and detection of the tracer in the outflow of a convective cloud at 12 km altitude which developed in the area of the dispersing Manaus pollution plume tagged with the tracer. The PMCH tracer was release from the top of a hotel west of the city center of Manaus, and sampled on board of HALO using adsorption tubes. The analysis of the PMCH tracer sampled with the tubes was conducted in the laboratory using a combination of a thermo-desorption system and a GC-MS with a detection limit of 1ppq (10$^{-15}$). The PMCH tracer could be clearly detected in the convective inflow and outflow with mixing ratios up to a factor of seven above atmospheric background values. The PMCH measurements are compared with observations of ambient tracer and used to infer loss and production of reactive nitrogen species and non-volatile and volatile aerosols during the convective transport.
Characterization of the northern Colorado front range tropospheric C2H6, CH4, HCHO, NH3, O3, and CO from ground based high spectral resolution FTIR measurements: oil & natural gas signature.

Early Career Scientist

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Abstract:

Over the past several years oil and natural gas (O&NG) related activities have increased tremendously in the northern Colorado Front Range. Fugitive emissions of gases from these activities impact air quality locally and have the potential to impact global air quality and climate. Consequently, extensive measurements of emitted atmospheric pollutants associated with O&NG activities have increased. However, the evolution and spatial distribution of multiple chemical species have mainly been characterized in limited field campaigns, i.e., FRAPPE and DISCOVER-AQ. In addition, recent studies show global reach of these emissions (e.g. Franco et al., 2016). In this work, we present long-term simultaneous measurements of tropospheric C2H6, CH4, HCHO, NH3, O3, and CO obtained from ground-based high spectral resolution solar absorption measurements in Boulder, Colorado from 2010 to present. The multi-year observations are used to identify the season to season changes in each species and distinguish short-term peak enhancements of C2H6 and CH4, likely caused by local O&NG point sources and NH3 from cattle feedlots. Our measurements, which are not influenced by the evolution of the planetary boundary layer, provide independent information to evaluate bottom-up emission inventories, as well as validate current and future satellite measurements, e.g., TEMPO.

6.119 Study of methane variability in Mexico City from total column and surface measurements.

Early Career Scientist

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Abstract:

Methane is one of the most important greenhouse gases due to its warming potential and fast growing increase in the atmosphere. Although the CH$_4$ concentration in the atmosphere and trend is well known on a global scale, the amounts attributed to specific local sources have large uncertainties and are of great relevance for chemical processes. For example, the emissions within megacities are not well understood and some studies suggest that CH$_4$ emissions in urban areas could be greater than previously estimated.

The total column of CH$_4$ and its surface concentration is being constantly measured South of Mexico City at the UNAM campus (N 19.32°, W 99.17°, 2260 m.a.s.l.). The total columns were retrieved from five years of ground-based solar absorption measurements with an FTIR spectrometer (VERTEX 80) and in situ measurements at the surface have been made with a commercial cavity ring-down spectrometer (Picarro G2401) for the past three years. In this work we present results from the evaluated data during this period in order to identify patterns and trends focusing on events with large concentrations. The co-location of both instruments gives us the opportunity to study the variability of the methane in a megacity using two different approaches.
6.120 Attribution of Atmospheric Sulfur Dioxide over the English Channel to Dimethylsulfide and Changing Ship Emissions.

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Abstract:
Atmospheric sulfur dioxide (SO$_2$) was measured continuously from the Penlee Point Atmospheric Observatory (PPAO) near Plymouth, United Kingdom between May 2014 and November 2015. This coastal site is exposed to marine air across a wide wind sector. The predominant southwesterly winds carry relatively clean background Atlantic air. In contrast, air from the southeast is heavily influenced by exhaust plumes from ships in the English Channel as well as near the Plymouth Sound. New International Maritime Organization (IMO) regulation came into force in January 2015 to reduce the maximum allowed sulfur content in ships’ fuel tenfold in Sulfur Emission Control Areas such as the English Channel. Our observations suggest a three-fold reduction in ship-emitted SO$_2$ from 2014 to 2015. Apparent fuel sulfur content calculated from coincidental SO$_2$ and carbon dioxide (CO$_2$) peaks from local ship plumes show a high level of compliance to the IMO regulation (>95%) in both years (~70% of ships in 2014 were already emitting at levels below the 2015 cap). Dimethylsulfide (DMS) is an important source of atmospheric SO$_2$ even in this semi-polluted region. The relative contribution of DMS oxidation to the SO$_2$ burden over the English Channel increased from ~1/3 in 2014 to ~1/2 in 2015 due to the reduction in ship sulfur emissions. Our diel analysis suggests that SO$_2$ is removed from the marine atmospheric boundary layer in about half a day, with dry deposition to the ocean accounting for a quarter of the total loss.
6.123 ORGANOCHLORINE PESTICIDES IN THE ATMOSPHERIC AEROSOL AROUND MEXICO COUNTRY USING PASSIVE SAMPLERS.

Early Career Scientist

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Abstract:

Organochlorine pesticides (OP) are part of persistent organic pollutants. They show toxic, carcinogenic and mutagenic properties. OP and their degradation products can be detected in the atmosphere long time after their emissions and far away to their sources, like in the case of pristine zones. To evaluate the OP presence in remote areas, passive samplers have been employed. In our study, we used passive samplers with polyurethane foam (PUF) to collect atmospheric aerosol in five sites around Mexico Country. Samplings were carried out during 90 days on 2014 (n=8) and 2015 (n=20). Samplings sites are part of the Network of Atmospheric Observatories (RUOA) from Universidad Nacional Autonoma de Mexico. PUF were located in a special homemade glass cell. Dichloromethane was used to extract the OP from the PUF using an ultrasound bath. Temperature, potency and time were controlled. According to factorial design (2^3), the best extraction conditions were 50 °C, 40 % and 40 min, twice. Endosulfan I was found in all sites, except at the east of the country (Los Tuxtlas, protected natural area 530 m.a.s.l.). Other OP in minor concentrations were 4,4'-DDE y β-HCH. Actually, we are analyzing 60 samples, we will show their seasonal and spatial trend of the found OP.
6.126 Inverse modeling of CH4 surface fluxes using GOSAT observations - Level 4 product updates.

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Abstract:

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Greenhouse gases Observing SATellite (GOSAT) Level 4 (L4) product v1.03 – monthly regional surface CH\textsubscript{4} flux estimates by inverse modeling from GOSAT XCH\textsubscript{4} and ground-based CH\textsubscript{4} observational data by WDCGG and NIES datasets now cover the 3-year period starting from June 2009. The L4 product extension till September 2013 was made using the recent version of EDGAR emission inventory and extension of GFED v3.1 fire emissions. The products provide opportunity to study the interannual flux variability including events of CH\textsubscript{4} emissions from large-scale climate anomalies and forest fires in Russia and Amazonia in 2010. With adding GOSAT retrievals to the flux estimation (called GOSAT-L4), we found enhanced fluxes in tropical Africa (17% from a priori and 10% from flux estimates using ground-based observations only, called SURFACE), tropical and subtropical South America (12% and 9% respectively), and East Asia (21% and 6% respectively), but lowered fluxes in South and Southeast Asia (12% and 14% respectively). In 2010 raging fires occurred in Brazil and Bolivia under severe drought, and the highest biomass burning fluxes in central part of South America were estimated in 2010 during the simulation period 2010-2012. The intensity of the 2010 biomass burning flux was enhanced in GOSAT-L4 compared with a priori of GFED v3.1 and SURFACE. In Russia, two fire events occurred in 2010 and 2012 under very hot and relatively dry condition. The 2010 fires occurred over European Russia, and a large departure from the GFED estimates was not shown in both SURFACE and GOSAT-L4. The GOSAT-L4 biomass burning fluxes were similar to a priori, but lower than SURFACE (particularly in the eastern part of Siberia).
Abstract:

An improved understanding of the variations in the mass concentration and size distribution of black carbon (BC) in the free troposphere (FT) over East Asia, where BC emissions are very high, is needed to reliably estimate the radiative forcing of BC in climate models. We measured these parameters and the carbon monoxide (CO) concentration by conducting the A-FORCE 2013W aircraft campaign in East Asia in winter 2013 and compared these data with measurements made in the same region in spring 2009. The median BC concentrations in the FT originating from North China (NC) and South China (SC) showed different seasonal variations, which were primarily caused by variations in meteorological conditions. CO concentrations above the background were much higher in SC than in NC in both seasons, suggesting a more active upward transport of water-insoluble CO. In SC, precipitation greatly increased from winter to spring, leading to an increased wet deposition of BC. As a result, the median BC concentration in the FT was highest in SC air in winter. This season and region were optimal for the effective transport of BC from the planetary boundary layer to the FT. The count median diameters of the BC size distributions generally decreased with altitude via wet removal during upward transport. The altitude dependence of the BC size distributions was similar in winter and spring, in accord with the similarity in the BC mixing state. The observed BC concentrations and microphysical properties will be useful for evaluating the performance of climate models.
Towards developing a climatology for assessing the radiative impact of smoke aerosols on the UV radiation.

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Abstract:

We have been working on the development of a new climatology of the UV radiation through a combination of measurements and modeling. The UV actinic flux spectrally resolved data have been collected during a number of field aircraft missions by the NCAR group. The actinic flux data are provided by the SAFS instruments, which were deployed in several airborne missions (including several AVE campaigns, TC4, INTEX-B, ARCTAS, ARCTAS, and SEAC4RS). The data are being used in conjunction with the modeling to identify the radiative signal of UV-active aerosol, especially smoke. Several different field campaigns are being analyzed. The modeling component includes the WRF-SMOKE model that has been developed by the Dr. Sokolik’s group. This model uses the satellite products to simulate the fire emission for a specific event at the high spatial and temporal resolution. Smoke is treated via the representative size distribution and refractive indexes to compute its optical characteristic. An approach to characterize the radiative impact of smoke aerosol has been developed and applied to estimate quantitatively the magnitudes of this impact. The effects of the smoke on the photolysis rates will be also presented.

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Abstract:

Ground level ozone continues to be a problem in the Eastern United States as well as East Asia. We present a compilation of recent studies demonstrating how photochemistry and meteorology conspire to generate high levels of secondary pollutants (ozone and aerosols) on regional and local scales. In situ observations from the surface and aircraft as well as remotely sensed data are used to constrain and improve numerical simulations performed with CMAQ and CAMx. We demonstrate improvements in emissions (vehicular NOx) chemical mechanisms (the removal of alkyl nitrates) and vertical transport (high resolution WRF meteorology) that bring model results closer to observations and provide guidance for abatement strategies in the Mid Atlantic States. These are constrained with the chemistry and meteorology of central China. Results provide insight into the most effective means of improving air quality.
Abstract:

The Arctic is a complex and poorly understood aerosol environment, impacted by strong anthropogenic contributions during winter to spring, and by regional sources in cleaner summer months. Our understanding of summertime Arctic aerosol and cloud remains incomplete, in part due to a scarcity of measurements focusing on the role of regional sources in shaping aerosol chemical and physical properties. To aid in addressing these uncertainties we made measurements of aerosol physical and chemical properties aboard an aircraft, as part of the NETCARE project, allowing measurements from 60 to 3000 meters over ice and open water. This summertime campaign was based in the Canadian High Arctic, at Resolute, NU (74°N), in a general time period and location that was shown to have high biological activity in the surface ocean. Here, we focus on observations of
submicron aerosol composition from an aerosol mass spectrometer. Under stable and clean atmospheric conditions with relatively low carbon monoxide and black carbon concentrations (< 100 ppb and < 50 ng/m³, respectively), we observe organic aerosol (OA)-to-sulfate ratios ranging from ~0.5 to greater than 6 with evidence for enhancement within the lower boundary layer. OA at lower altitudes tended to be less-oxygenated, with lower O-to-C and higher H-to-C ratios, compared to OA observed aloft. Methanesulfonic acid (MSA), a marker for the contribution of ocean-derived biogenic sulphur, was also observed in submicron aerosol. MSA-to-sulfate ratios ranged from near zero to ~0.3 and tended to increase within the lower boundary layer, suggesting a contribution to aerosol loading from the ocean. While there are contributions from both primary and secondary aerosol across the size distribution, in some cases enhanced concentrations of OA and MSA were associated with aerosol growth. With these observations we explore the composition and formation processes contributing to cloud condensation nuclei in the summertime Arctic.
**6.135 CO2 variability and trends in Mexico.**

Early Career Scientist

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**Abstract:**

Carbon dioxide has been rising since the Industrial Era to levels over 400 ppm in the last years. CO2 is the most important greenhouse gas from anthropogenic sources, it has a long atmospheric lifetime which makes for its high background atmospheric concentration that shows relatively small spatial and temporal variations. To account for this variability, dense and continuous CO2 observations are needed. Since 2014, CO2 has been monitored in six stations from the University Network of Atmospheric Observatories RUOA (www.ruoa.unam.mx) using commercial cavity ring-down spectrometers. Three of these stations are within protected natural areas and are considered remote sites, whereas the rest are located within urban areas. At two of these sites, Altzomoni (N19.18°, W 98.65, 3985 masl) and the UNAM campus in Mexico City (N 19.32°, W 99.17°W, 2260 masl), the total vertical column of this gas is determined from solar absorption infrared measurements made in the near-infrared (NIR) spectral region and compared to satellite observations. In this work we present and interpret the data collected at these stations and show the seasonal variability and trends of CO2 in very distinct regions of the Mexican territory.
Atmospheric studies in large cities are of great relevance since pollution affects air quality and human health. According to the latest revision of the World Urbanization Prospects released by the Department of Economic and Social Affairs of the United Nations Secretariat, in 2014 about 80 per cent of the Latin American population lived in urban areas. Furthermore, over the coming decades urbanization is expected to increase in all regions of the world. Being the Mexico City Metropolitan Area (MCMA) one of the largest urban areas of the world with around 21 million inhabitants, to improve air quality monitoring of the MCMA atmosphere becomes a pressing matter.

A network of Multi Axis Differential Optical Absorption Spectrometers (MAX-DOAS) has been established in strategic sites within the MCMA. Four instruments are now in operation with the aim to study the variability and spatial distribution of key pollutants, providing results of nitrogen dioxide (NO$_2$), formaldehyde (HCHO) and the oxygen dimer (O$_4$) slant column densities (SCDs). A numerical code has been written to retrieve gas profiles of NO$_2$ and HCHO using radiative transfer simulations and to derive vertical column densities (VCDs). Preliminary results of the variability of VCDs of these trace gases in the atmosphere of the MCMA are presented, which will bring new insights to the current knowledge of the abundances of these molecules, transport patterns, emissions as well as the frequency and origin of extraordinary events. These studies complement surface measurements routinely conducted by the air quality monitoring network of the MCMA, providing information on the vertical distribution of the studied species as well as the total column amount within the boundary layer. In addition, these measurements are useful to validate current and future satellite observations such as OMI, TROPOMI and
TEMPO.
6.139 Downmixing of stratospheric air observed from FTIR measurements in central Mexico.

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Abstract:

A new tropical ground-based station part of the Network for the Detection of Atmospheric Composition Change (NDACC) is operating in Mexico since 2012. Altzomoni (19.12° N, 98.66° W) is located at an altitude of 3,985 m a.s.l. within a national park in central Mexico next to the Popocatépetl and Iztaccíhuatl volcanos. The station contributes to NDACC with high resolution solar absorption measurements by means of a Fourier Transform InfraRed (FTIR) spectrometer, from which vertical profiles and total column of several atmospheric species such as O₃, HNO₃, HCl, HF, CO, N₂O and CH₄ are retrieved. The station offers the opportunity not only to contribute to the scarce information available for the annual variability, trends and vertical distribution of important gases at these latitudes, but also to study the exchange processes between the stratosphere and the troposphere.

In this contribution we will show some examples of events in which the vertical downmixing of stratospheric air is detected. Time series in which O₃ and HCl present anti-correlations to tropospheric gases such as CH₄ and N₂O are shown. The potential temperature plot shows that these are periods of strong instability and that the Tropical Tropopause Layer (TTL) is frequently breaking allowing an exchange into lower altitudes. Discussion of the occurrences and frequency of these events and their implications will be included.
6.142 Satellite measurement of the lower tropospheric ozone enhancement over East Asia - elimination of the effect from the upper troposphere and lower stratosphere.

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Abstract:

The lower tropospheric ozone enhancement over Central and Eastern China (CEC) was reported by Hayashida et al. (2015) by using the Ozone Monitoring Instrument (OMI) multiple-layer product retrieved by Liu et al. (2010). The lower tropospheric ozone distribution maps were first obtained from the UV space-borne measurements by their study. However, to clarify the enhancement in the concentration of the lowermost ozone, it is necessary to eliminate the effect of ozone variation in the upper troposphere and lower stratosphere (UT/LS) because of large smoothing errors in the retrieval scheme. In this study, a scheme was developed to eliminate the artificial effect of UT/LS ozone enhancement on lower tropospheric ozone retrieval. By applying the screening scheme, we could remove the effect of the UT/LS ozone enhancement on the lower tropospheric ozone and show a clear enhancement in the lower tropospheric ozone over CEC in June 2006 and confirmed the conclusion derived by Hayashida et al. (2015).

We also compared satellite measurements with model simulations for \( \text{O}_3 \) by using a global chemical transport model MRI-CCM2 (Deushi and Shibata, 2011) and a regional model NHM-Chem (Kajino et al., 2012). Those model simulations could well reproduce the observed \( \text{O}_3 \) enhancement over CEC in June 2006. The effects of emissions from the open crop residue burning (OCRB) in the North China Plain on lower tropospheric ozone were also examined by utilizing the emission inventory developed by K. Yamaji (Yamaji et al., 2010). The difference between the \( \text{O}_3 \) with and without the OCRB effect revealed that the effect of OCRB on \( \text{O}_3 \) enhancement is not critical.

References

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Yamaji K et al. (2010): ACP, 10 (15):7353-7368.
6.143 Characteristics and transformations of pollution gases to particulate matters in fog-haze episodes.

Early Career Scientist

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Abstract:

There was a multi-day widespread fog-haze processes during 4 and 12 June 2012 in Huaihe River Basin of China. In the process of continuous 9 d fog-haze episode, there was a moderate haze - heavy haze - fog - severe haze - moderate haze conversion process, and also the pollution gases transformed to particulate matter phenomenon. During the haze-fog process, the visibility reduced with the wind speed decrease, and lifted with the wind speed increase. Lower wind speed converge the pollutants, improve the aerosol particle nucleation condensation and condensation growth, thereby reducing visibility. The formation and development of moderate and severe haze were accompanied with increasing specific humidity, while the haze-fog conversion processes were accompanied with decreasing specific humidity. The higher the specific humidity, the longer the fog-haze duration. The higher the specific humidity before the fog, the lower the minimum visibility during the fog.

Based on the same site, Visibility was positively correlated with the concentration of SO$_2$ (Pearson correlation coefficients is 0.42), but was inversely correlated with PM$_{2.5}$ and PM$_{10}$ concentrations(Pearson correlation coefficients are -0.49 and -0.56, respectively). On the contrastive analysis between the study site and its upwind site, all types of pollutant concentrations have the same variation trends, and the upwind ones earlier than those of the study site. During the fog processes, the upwind PM$_{10}$ and PM$_{2.5}$ concentrations lower than those of study site. For the contaminated gas, the CO concentration has a small variation, and the upwind SO$_2$, NO$_2$ concentrations were higher than about 2 times those of downwind. However, the downwind NO and PM$_{10}$ concentrations were about 1.5 times higher than those of the upwind. The pollutants conversions may be occurred during the fog processes.
6.145 Observations of atmospheric black carbon mass concentrations from East Asia to the open oceans: Constraining emission strengths and wet deposition rates.

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Abstract:

Both emission strengths and removal rates of black carbon (BC) particles need to be known to elucidate their atmospheric behavior and impact on the Earth’s climate. However, they are still poorly characterized. We selected COSMOS and SP2 instruments as reliable instruments and conducted long-term observations at remote sites in Asia and over the Arctic/Pacific/Indian Oceans using R/V Mirai of JAMSTEC, to provide unique information on wet removal rates and the emission strengths of important source regions (China and others). Here we focus on the long-term (2009–2015) measurements at Fukue Island, western Japan (Kanaya et al., ACPD, 2016), receiving outflow from the major source regions within 6–46 hours. Wet removal of BC was clearly suggested from decreasing trend in the observed $\Delta$BC/$\Delta$CO ratios against the accumulated precipitation along backward trajectories (APT) for the last 3 days. The remaining fraction of BC in the atmosphere was fitted reasonably well by a stretched exponential decay curve against APT; a single set of fitting parameters was sufficient to represent the results for air masses originating from different areas. An accumulated precipitation of 15.0 ± 3.2 mm halved the BC mass concentration. BC/CO emission ratios estimated from cases with zero APT (5.2–6.9 ng m$^{-3}$ ppb$^{-1}$) varied over the air mass origin areas. The significantly higher BC/CO emission ratios adopted in the Regional Emission inventory in Asia (REAS) version 2 (8.3–23 ng m$^{-3}$ ppb$^{-1}$) over CEC and Korea needed to be reduced at least by factors of 1.3 and 2.8, respectively, but the ratio for Japan was reasonable. After correcting for the wet removal effect, trends in the BC mass concentrations were almost flat for the air masses from CEC and Korea and decreasing for those from Japan during 2009–2015. This long-term data set will provide a benchmark for testing regional/global-scale model simulations.
6.146 Trace gas transport out of the Indian Summer Monsoon.

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Abstract:

The trace gas transport out of the Indian summer monsoon was investigated as part of the aircraft campaign OMO (Oxidation Mechanism Observations) using the German research aircraft HALO (High Altitude and Long Range Research Aircraft) in July/August 2015. HALO was based alternatively at Paphos/Cyprus and Gan/Maledives. Flights took place over the Mediterranean Sea, the Arabian Peninsula and the Arabian Sea. Here we investigate the distribution of carbon monoxide (CO) and methane (CH4) in the upper troposphere, measured in-situ with the IR-laser absorption spectrometer TRISTAR. During OMO enhanced concentrations of methane and carbon monoxide were detected in the Asian Summer Monsoon Anticyclone at altitudes between 11km and 15km. Mixing ratios exceeded background levels for CO and CH4 by 10-15ppb and 30-40ppb, respectively. The enhancement in the CO concentration falls within the range of tropospheric variability, while the methane enhancement is much higher than its natural variability. Therefore methane is a very good tracer for air masses influenced by the monsoon. This is confirmed by back trajectory calculations with FLEXPART, indicating convective transport from India approximately 10 days before the observations. A comparison of observations with EMAC model simulations (global atmosphere-chemistry model ECHAM/MESSy) generally agree within ± 5.5 % and ± 1.4 % for CO and CH4, respectively.
6.148 Atmospheric chemistry of reactive nitrogen species over Indo-Gangetic plains (India).

Early Career Scientist

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Abstract:

In India around 70 % of population lives in villages taking care of agriculture to meet the demand of food supply. In order to get higher yield of agriculture and food product, increased practice of fertilizer application has added extra load of nutrients especially the reactive nitrogen (Nr) species viz NH$_3$ and NO$_x$. Growing energy demand has resulted in increased emission of NO$_x$ from coal combustion in thermal power plant and the petroleum combustion in transport sector. In addition, biomass burning in traditional cooking and heating is also a significant source of NO$_x$ in Indian region. Significance of the study lies in the fact that increasing Nr emissions have adverse impact on human health, plant, soil and water bodies directly and to see the effect, knowledge of emission and deposition for Nr at different sites should be there.

Considering the implications of these two species (NH$_3$ and NO$_x$) in changing N cycle, the present study was carried out in Indo-Gangetic plains (IGP) at two sites of different characteristic (urban and rural) to study the emissions and atmospheric levels in relation to their sources and role of meteorological parameters. Study presents seasonal and diurnal variations of gaseous reactive nitrogen species at sites to observe the contribution of different sources of atmospheric Nr. Average concentrations of NH$_3$ at urban and rural site have been recorded as 40.4 ±16.8 and 51.57 ±22.8 µg/m$^3$ respectively. The average concentrations of NO$_2$ have been recorded as 24.4 ±13.5 and 18.8 ± 12.6 µg/m$^3$ at urban & rural site respectively. Dynamics of Nr will be discussed in details at the conference.

Key words: Reactive nitrogen, ammonia, nitrogen dioxide, agriculture
**6.149 Source Apportionment of PM2.5 and Secondary Organic Aerosol Estimation in Pearl River Delta of China.**

Early Career Scientist

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Abstract:

PM$_{2.5}$, causing decrease of atmospheric visibility and adverse health effects, becomes the focus of air pollution control in China. Pearl River Delta (PRD) is one of the most economically developed regions in China and ever experienced severe air pollution. However, the air quality in PRD got better and better after strict air pollution control measures applied and successful economic transformation in the recent ten years, with PM$_{2.5}$ finally meeting the national standard in 2015. PRD is now regarded as a model of making a balance between development and environment. In this study, the latest PM$_{2.5}$ source information in PRD was explored, with the three cities around the estuary of PRD, i.e., Dongguan, Shenzhen, and Zhuhai, selected as typical sampling domains. There were five sampling sites setup in each city and all the PM$_{2.5}$ samples were collected in the four seasons from 2014 to 2015. Water-soluble inorganic ions, organic carbon (OC), elemental carbon (EC), and trace elements were detected and quantified for these samples. The positive matrix factorization (PMF) model was applied to the PM$_{2.5}$ dataset in each city to
make source apportionment. The results show that: (1) the mean mass concentration of PM$_{2.5}$ was 41.4 μg/m$^3$ in the three cities, far lower than ten years before; (2) secondary sulfate (21-30%), vehicle emissions (15-27%), and secondary nitrate (10-12%) were the main sources of PM$_{2.5}$. Dust, biomass burning, ships, sea salts, and industrial dust each contributed 1.6-8.2%; (3) the analysis of spatio-temporal variation of main sources of PM$_{2.5}$ indicated that vehicle emissions mainly came from local sources, secondary sulfate mainly came from regional formation, while both local and regional sources contributed significantly to secondary nitrate; (4) secondary organic aerosol (SOA) was also identified and estimated by the PMF model and found to account for 34% of total organic mass on average.
6.150 Convective transport of short-lived hydrocarbons and bromocarbons from the surface to the upper troposphere and lower stratosphere.

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Abstract:

Deep convection is the primary mechanism that delivers important chemical trace gases into the upper troposphere and the lower stratosphere (UT/LS), where they exert significant impacts on atmospheric O$_3$ and OH. The Western Pacific has been viewed traditionally as the primary convective lofting region for air to enter the tropical tropopause layer (TTL), although deep convective injection over the continental U.S. has also been proposed as a potential pathway. We have analyzed aircraft measurements of CO$_2$, NMHCs (CO, ethyne, ethane, HCHO), and very-short-lived bromocarbons from multiple aircraft missions with trace gas simulations from the NASA GOES-5 chemistry climate model. The results from the NASA SEAC$^4$RS mission over the N. American continental sub-tropics are compared with those from the NASA ATTREX and NSF CONTRAST missions over the Western Pacific to examine the transport timescale and transport efficiency of these chemical compounds from the surface to the UT/LS and to investigate how convective transport differ in these two regions. We found that the Western Pacific is convectively efficient with rapid vertical transport from the surface to the 365K potential temperature level (~16 km) within 60 days. Convective transport over the continental N. America is less frequent and much slower (120 days from surface to 365K), but sporadic injection can be as high as the 400K potential temperature level (~18 km) during some occasions. We will extend the analysis to additional aircraft missions, including the NASA TC$^4$ mission in the deep tropical Central America and the early summer NSF DC$^3$ mission over the continental U.S, to achieve a better understanding of convective transport into the UT/LS and how it varies with season and region.
6.151 Formaldehyde column density measurements as a suitable pathway to estimate near-surface ozone tendencies from space.

Early Career Scientist

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Abstract:
In support of future satellite missions, NASA’s DISCOVER-AQ field campaign was designed to enable exploration of relationships between column measurements of trace species relevant to air quality at high spatial and temporal resolution. Because CH$_2$O has a very short lifetime, its vertical distribution is heavily weighted towards the surface and any correlation between CH$_2$O and O$_3$ has the potential to be useful in estimating near-surface O$_3$ tendencies from space. Initial analysis of DISCOVER-AQ data showed a modest correlation between column measurements of CH$_2$O and O$_3$, and further analysis revealed distinct behavioral differences in the O$_3$-CH$_2$O relationship between data collected in Maryland and Houston. In Maryland, we found that the O$_3$-CH$_2$O relationship was strongest when there was high temporal variability in CH$_2$O column densities. In Houston, on the other hand, approaching the dataset from a spatial perspective revealed interesting behavior and suggested that time-dependent transport of emissions from the ship channel area obfuscated the observed O$_3$-CH$_2$O relationship. In Maryland, a strong diurnal pattern in biogenic emissions caused CH$_2$O to, in general, increase monotonically throughout the day. In Houston, however, anthropogenic emissions from the ship channel dominated the local hydrocarbon environment, and no discernable diurnal trend in CH$_2$O was observed. Box model simulations suggest that ambient CH$_2$O mixing ratios have a weak diurnal trend unless concurrent changes in the local hydrocarbon oxidation environment occur. When a diurnal trend in isoprene was introduced to model simulations, co-variance between O$_3$ and CH$_2$O was predicted, which is in agreement with our observations. While additional measurements and studies involving 3D models would be necessary to validate these results and determine the regional applicability of the O$_3$-CH$_2$O relationship, the results presented here provide compelling insight into the ability of future satellite missions to aid in monitoring near-surface air quality.

Early Career Scientist

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Abstract:

The distribution of most trace species within the troposphere depends on a complexity interplay between transport and chemical loss. Holzer and Waugh (2015) recently introduced the concept of a path-dependent lifetime, $\tau_{C}(r)$, that parameterizes the integrated chemical loss during transport to a given location $r$. Here we present estimates of path-dependent lifetimes of a suite of CFC replacement gases based on surface and aircraft observations. Chemical transport model simulations are then used to investigate the characteristics of $\tau_{C}$, its linkage to tropospheric transport and OH abundance. These simulations suggest that observational estimates of $\tau_{C}$ may place useful constraints on the tropospheric OH.
Understanding the origin of CCN in the remote troposphere.

Early Career Scientist

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Abstract:

The ability to fully define aerosol-cloud-climate interactions requires an understanding of the budget of primary (emitted) and secondary (produced in situ) cloud condensation nuclei (CCN) in different regions and factors that control changes in their abundance. Aerosol indirect radiative forcing, which is controlled by CCN abundance, is the largest uncertainty in climate forcing.

Global earth system models incorporate aerosol microphysics to describe the evolution of size-resolved aerosol properties that determine CCN abundance. One of the most important parameters contributing to CCN uncertainty in these models is the aerosol size distribution. Specifically, the uncertainty in the diameter of the Aitken mode accounts for 40% of model variance in CCN abundance in remote oceanic regions. Dry deposition of the accumulation mode contributes 30% to model variance. These findings clearly point to the need for improved understanding of aerosol production mechanisms, sinks and transformations especially in the remote troposphere. Only if these processes are understood can models evaluate the global CCN budget and thus the role of CCN in controlling cloud radiative properties, lifetime and extent.

The Atmospheric Tomography (ATom) airborne research experiment will sample the remote troposphere over the Atlantic and Pacific basins with near-pole-to-pole coverage in July and August 2016. A total of four such global circuits will be made, one in each season. These flights will scan the atmosphere in continuous ascents and descents between 0.2 and 12km, giving both the latitudinal and vertical distribution of the aerosol.
Here we present preliminary results of the first deployment. Dry particle size distributions between ~ 65 to 1000 nm in diameter will be measured with a suite of fast-response size distribution instruments. Preliminary results should include the spatial variation in the abundance of CCN-sized particles and their association with regions of new particle formation and with long-range transport from continental sources.
6.155 Influence of the weather pattern on the photochemical ozone production of Lima, Peru.

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Abstract:
Radiosondes launched in Lima (77ºW, 12ºS) since 1973 to 2014 at 7:00 am (local time), distinguish a period, between January and April, where surface inversions are more abundant due to the displacement of the semi-permanent high-pressure system to high latitudes. On the other hand, during winter and spring, the strengthening of the anticyclone leads to increasing subsidence inversions over Lima. The high-pressure condition also favors the upwelling of the cold Humboldt Current, whose interaction with the air generates a thick layer of cold and humid air, which covers the city. Therefore, this stratus cloud deck inhibits the thermal inversions due to the lack of solar radiation able to reach the surface.
In general terms, the weather pattern constrains the strong photochemical ozone production within a couple of months. Also, the precursors deposition seems to keep limited the ozone concentration as well. In 2010, the official monitoring network was established in Lima, currently an attainment area, where levels of ~50 ppbv (MDA8) are often reached in summer especially in downwind locations. VOC (C6-C12) campaigns performed in 2015 to complement existing surface observations, also indicate to the northern and northeastern part of the city as locations with relatively high photochemical production. For instance, the low ratios xylene:benzene reached at noon, show the impact of aged air masses arriving from upwind locations during this period: 5 (7:00) / 3 (12:00) / 5 (18:00). Further, during this year photochemical campaigns and atmospheric structure analysis will be conducted to gain insight into this upwelling region.
6.156 Ozone responses to atmospheric modulations in Malaysia.

Early Career Scientist

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Abstract:

In the tropics, changes in ambient temperature and daylight hours are less pronounced compared to higher latitudes where the summer and winter seasons show large fluctuations in weather. However, ambient ozone (O\textsubscript{3}) concentrations in the tropics can still be modulated by cyclical atmospheric processes such as the Asian monsoon and El Niño Southern Oscillation (ENSO) which influences cloud cover and rainfall patterns. This study attempts to determine the relative influence of atmospheric modulations on ambient O\textsubscript{3} concentrations in Malaysia, a country located within 7° North of the equator. Long term O\textsubscript{3} and meteorological parameters data (>10 years) from a network of ambient air quality monitoring sites made available by the Department of Environment Malaysia is analysed. Preliminary statistical analysis results on five selected stations (1997 - 2011) showed that daily maximum O\textsubscript{3} concentrations were not spatially consistent to ENSO and monsoonal modulation indicating stronger localised influence on O\textsubscript{3} concentrations. Spectral analysis results showed a shift from annual to semi-annual cycle moving from north to south of the Malaysian peninsula indicating sensitivity to the shift in Intertropical Convergence Zone. The current work is expected to provide more conclusive evidence on the relative influence of atmospheric modulations to ambient O\textsubscript{3} concentrations in the tropics.
Chemical four dimensional variational data assimilation in WRFDA-Chem: Improving Black Carbon Emission Inventories during ARCTAS-CARB.

Early Career Scientist

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Abstract:

Wildfires contribute significantly to regional haze events globally, and they are potentially becoming more commonplace with increasing droughts due to climate change. PM2.5 emissions from wildfires are highly uncertain, with global annual totals varying by a factor of 2 to 3 and regional rates varying by up to a factor of 10. At the high resolution required to predict PM2.5 exposure events, this variance is attributable to differences in methodology, differing land cover datasets, spatial variation in fire locations, and limited understanding of fast transient fire behavior. Here we apply an adjoint-based online chemical inverse modeling tool, WRFDA-Chem, to constrain black carbon aerosol (BC) emissions from fires during the 2008 ARCTAS-CARB field campaign. Although BC comprises only about 5% of PM2.5 from fires during that campaign, it is one of the simpler aerosol constituents to model and measure. The constrained emission fields on June 22, 2008 are evaluated through cross validation with surface and aircraft observations on subsequent days. We find that over-predictions of BC in southern California are caused by high bias in coastal fire emissions between Los Angeles and San Francisco and also in anthropogenic inventories in both of those cities. Low emission bias exists in areas where smoke coverage might inhibit satellite detection of fire hot spots. We identify several weaknesses in the temporal distribution of emissions, including a missing early morning emission peak associated with local, persistent, large-scale forest fires. Diurnal studies of different fire types should be prioritized in future campaigns in order to improve fire inventories on the temporal and spatial scales required to predict PM2.5 exposure. WRFDA-Chem will provide insights to reduce uncertainties in aerosol and trace gas emission inventories, which feed into air quality monitoring and health and climate impact studies.
Abstract:

Nitrogen oxide radicals (NO\textsubscript{x} = NO + NO\textsubscript{2}) play a central role in controlling the oxidizing capacity of the troposphere, and peroxyacetyl nitrate (PAN) serves as their chief reservoir. Observations of PAN are notoriously difficult to simulate in models because the stability of PAN depends strongly on vertical transport, and its formation is strongly influenced by non methane volatile organic compounds with uncertain emissions and complex oxidation chemistry. New observations of PAN from the Tropospheric Emission Spectrometer (TES) offer an unprecedented way to assess the spatial distribution, seasonality, and interannual variability of this species in the free troposphere. We analyze observations over North America using a combination of tools including backward trajectories and the GEOS-Chem chemical transport model to diagnose the sources of PAN as well as model deficiencies and their implications for free tropospheric oxidant distributions.

We plan to primarily focus our discussion on summer, when previous aircraft observations from multiple campaigns have suggested there is a reservoir of 200–400 pptv PAN between 4 and 8 km over northern midlatitudes. The new TES data does show an increase in PAN from spring to summer in the free troposphere over remote regions. We find no systematic differences in the distributions of PAN when retrievals are segregated by either the underlying NO\textsubscript{x} emissions, or the presence of smoke in the atmospheric column. The data also demonstrates that the model overestimates PAN in the free troposphere over the southeastern U.S. Through an analysis of the enhancement ratio of PAN relative to simultaneous TES CO measurements, we find that the model is likely to systematically underestimate the contribution of fires to elevated PAN during summer months over western North America.
Measurements of black carbon containing aerosols in South Korea during KORUS-AQ.

Early Career Scientist

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Abstract:

Black carbon (BC) is a light-absorbing aerosol with strong anthropogenic sources that has important climatic and health impacts, both regionally and globally. Materials internally mixed with BC, including water, affect its optical properties and lifetime in the atmosphere, and thus are critical to understanding its ultimate impacts; these processes are still highly uncertain. The NASA KORUS-AQ campaign during the summer of 2016 was a multi-platform research campaign focusing on studying air quality over South Korea, in a region with particularly high BC emissions and loadings. The NOAA Humidified-Dual Single Particle Soot Photometer (HD-SP2) was deployed on the NASA DC-8 aircraft to measure the refractory BC content and mass mixing ratio (MMR) of single BC-containing aerosol particles sampled in situ under dry and humidified conditions. These measurements facilitated the characterization of water uptake by black carbon-containing particles, and improved the understanding of BC-containing aerosol’s ambient properties and evolution in the atmosphere. Preliminary results from the KORUS-AQ field campaign are discussed, including BC MMRs, degree of internal mixing, and propensity to be associated with water-active materials.
Observations of particle organic nitrate from airborne/ground platforms: Insights into vertical/geographical distribution, gas/particle partitioning, losses, and contribution to total particle nitrate.

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Abstract:
Organic nitrate formation in the atmosphere represents a sink of NO$_X$ and termination of the HO$_X$/NO$_X$-ozone formation cycles, can act as a NO$_X$ reservoir transporting reactive nitrogen, and contributes to secondary organic aerosol formation. However, particle-phase organic nitrates (pRONO$_2$) are rarely measured and thus poorly understood. We use simultaneous measurements of pRONO$_2$ and of total (gas+particle) organic nitrate...
(totRONO$_2$), organic aerosols (OA), and ammonium nitrate from the DC3 and SEAC$^4$RS aircraft and several ground campaigns to investigate vertical/geographical distributions, gas/particle partitioning, losses, and contributions to total particle nitrate (pTotNO$_3$) over North America. Apportionment and quantification with aerosol mass spectrometry is evaluated. The fraction of pTotNO$_3$ that is organic increases steeply with decreasing pTotNO$_3$, approaching 100% at low pTotNO$_3$, primarily at rural/remote locations. pRONO$_2$ and totRONO$_2$ concentrations show strong vertical gradients, with a steep decrease from the top of the boundary layer (BL) up through the residual layer. However, pRONO$_2$ was 10-30% of totRONO$_2$ with little vertical gradient in gas/particle partitioning from the BL to upper troposphere (UT). pRONO$_2$ contribution to OA shows a moderate increase with decreasing OA in the BL and free troposphere (~2-3% by mass of nitrate group) with higher contributions at the lowest OA (5-8%), mostly observed in the UT. In the BL, RONO$_2$ gas/particle partitioning shows a trend with temperature, with higher particle-phase fraction at lower temperatures, as expected from partitioning theory. However, the temperature trend is much weaker than for single compound partitioning, which may be due to a broad mixture of species. Little to no dependence of pRONO$_2$/OA on RH or estimated particle water was observed in the BL, suggesting that losses of pRONO$_2$ due to hydrolysis are too rapid to observe in this dataset and there may be a substantial fraction of pRONO$_2$ species that are not prone to rapid hydrolysis.
6.168 Emission sources of C2-C7 non-methane hydrocarbons at an urban site of western India..

Early Career Scientist

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Abstract:

The mixing ratios of non-methane hydrocarbons (NMHCs) were measured at an urban site of Udaipur (26.58°N, 73.68°E, 598 m amsl) in western India. The air samples were collected during February 2015-January 2016 and analyzed for many NMHCs using a thermal desorption-gas chromatograph-flame ionization detector (TD-GC-FID) system. The mixing ratio of C2-C7 NMHCs show strong diurnal variation with peak during morning and evening rush hours for most of the species. In the winter season, the average mixing ratios of aromatic compounds benzene (C6H6) and toluene (C6H7) during morning hours were 1.6±0.1 ppbv and 2.5±0.4 ppbv, respectively. In the monsoon season, the NMHCs do not show significant diurnal dependence. Most of NMHCs show strong seasonal variation with highest during winter season and lowest during monsoon season. The seasonal change in long-range transport, boundary layer height and OH concentrations seem to contribute in the seasonal variations of NMHCs. The mixing ratio of isoprene shows correlation with solar flux and temperature in indicating its emission from biogenic sources. The observed NMHCs showed good correlation with each other in winter (r² > 0.5) suggesting their emissions from common or co-located sources. The emission of natural gas and use of liquid petroleum gas (LPG) contribute to elevated levels of ethane and propane. While emissions from vehicular exhaust are the dominant sources of ethene, propene, acetylene, benzene and toluene.
6.169 Long term trends in atmospheric composition from Australian ground-based remote sensing measurements.

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Abstract:

Remote sensing measurements of spectra in the mid infrared from a high resolution Fourier Transform Spectrometer (FTS) have been routinely collected from the campus of the University of Wollongong (UoW, lat -34.41, long 150.88E, alt 30 m) over a 20 year period. This measurement programme at UoW is part of the world-wide Network for the Detection of Composition Change (NDACC), a global network of stations using a range of remote sensing and in-situ measurement techniques. The mid-Infrared spectra recorded with the FTS, by its very nature captures the absorption information in the column from the ground to the top of the atmosphere. The spectra contain absorption features from many gases, most of which have some importance to chemistry and transport in the stratosphere (O3, HCl, ClONO2, HF), and troposphere (CO, HCN, NH3, CH4, CFC’s, OCS, ...).

The physics of the spectral absorption is such that there is some vertical information inherent in the spectra, depending on where the particular species resides in the atmosphere and its concentration. The UoW dataset, like many within the NDACC, therefore contains a very valuable long term record of a range of processes throughout the atmosphere. This dataset has been recently analysed in a methodical and consistent manner, using NDACC recommended analysis procedures. This data will be discussed and important features highlighted that demonstrates its current and future contributions to the understanding of quite broad scientific questions covering air quality, long range transport of pollutants, the importance of volatile organic compounds (for example HCHO), and trends in stratospheric Cl and its impact on O3.
6.172 Global synthesis of multi-year cloud condensation nuclei observations.

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Abstract:

Cloud condensation nuclei (CCN) influence the microphysical and optical properties of clouds. To predict cloud radiative properties, understanding the spatial and temporal variability of CCN concentrations in different environments is important. However,
currently, the effects of atmospheric particles on changes in cloud radiative forcing are still the largest contribution to uncertainty in climate forcing prediction (IPCC, 2013). Numerous field campaigns have explored detailed characteristics of CCN in many locations around the world. However, these short-term observations can generally not address seasonal or inter-annual variations, and comparison between sites is difficult. Here we present results of multi-year CCN number concentrations, as well as size distribution and chemical composition data covering at least one full year between 2006 and 2014. The 12 locations include ACTRIS stations (http://www.actris.net/) in Europe, and further sites in the Americas and Asia. These sites allow for temporal and spatial characterization of CCN variability in different atmospheric regimes. Covered environments include marine, remote-continental, boreal forest, rain forest, Arctic and monsoon-influenced environments, as well as boundary layer and free tropospheric conditions. Geometric mean diameters as well as the activation ratios on the basis of particles > 50 nm vary strongly among sites and throughout the seasons. For example, the rural-marine and rural-continental sites exhibit similar CCN concentration with little variation in the annual cycle, whereas in the boreal environment the annual cycle is more pronounced. Additionally, we investigate the persistence of CCN-concentrations over different timescales to explore meaningful averaging periods for global modelling of CCN. We find three different regimes: (1) CCN concentrations persist for a week or longer showing also seasonal patterns. (2) CCN concentrations persist for less than one week, and there is little seasonal pattern. And (3) CCN concentrations are highly variable and do not persist for longer than two days, however do show seasonal cycles.
6.174 Influence of meteorological and trace gas factors on surface ozone concentrations in the interior of South Africa.

Early Career Scientist

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Abstract:

South Africa’s own monitoring and modelling of surface ozone (O\textsubscript{3}) can be traced back to the early 1980s, however there are still significant gaps in knowledge about ozone in South Africa and of the conditions that contribute towards its formation. Accurate modelling of surface ozone within a local and regional context remains a challenge. This study was conducted to investigate the behaviour of O\textsubscript{3} concentration in surface air with several known meteorological and trace gas variables, using statistical methods to quantify the impact of these variables. This analysis focused specifically on the interior of South Africa where datasets from four inland monitoring locations were used. Multiple linear regression was applied to approximate the linear relationships between several predictor variables and the dependent variable O\textsubscript{3}. The coefficient of determination, R\textsuperscript{2}, for the prediction of the concentration of O\textsubscript{3} was found to be the highest when including the following variables: temperature, solar radiation, relative humidity, wind speed, wind direction, carbon monoxide and nitrogen oxides gas concentrations. Multicollinearity was evaluated in the model and determined to be low. In many O\textsubscript{3} prediction models, air temperature has been found to be the strongest single predictor of O\textsubscript{3} concentration. However, at three of the four monitoring sites studied, relative humidity appears to be the most important atmospheric factor influencing the variation in O\textsubscript{3} levels. Temperature and O\textsubscript{3} precursors (nitrogen oxides and carbon monoxide) were found to be the next significant variables. Relative humidity is important because it affects the amount of liquid water that condenses on the aerosol particles and the rate of reactive nitrogen loss to the particle phase. Several possibilities are being investigated to explain
the predominant influence of relative humidity on O$_3$ which could have implications for modelling tropospheric O$_3$ formation and destruction processes in this region of the world.
6.176 Intra-seasonal variability in wintertime aerosols at middle Indo-Gangetic Plain.

Early Career Scientist

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Abstract:

A unique wintertime aerosol behavior was identified at middle Indo-Gangetic Plain (IGP) with characteristic episodic variations in their mass loadings, physico-chemical properties and sources within winter months. To assess wintertime aerosol characteristics over middle IGP, near surface PM$_{10}$, PM$_{2.5}$ and their ionic compositions were examined. Concurrently columnar aerosol distribution through satellite measurements was also made. Exceptionally high PM$_{10}$ (268±107µgm$^{-3}$) and PM$_{2.5}$ (150±89µgm$^{-3}$) values were reported for the study period. The varying aerosol distribution during winter recognized a high aerosol loading episode (HALD; PM$_{10}$: 366±101µgm$^{-3}$, PM$_{2.5}$: 230±86µgm$^{-3}$), two medium aerosol loading episodes (MALD$_1$: PM$_{10}$: 272±86µgm$^{-3}$, PM$_{2.5}$: 171±70µgm$^{-3}$) & MALD$_2$: (PM$_{10}$: 230±41µgm$^{-3}$, PM$_{2.5}$: 123±51µgm$^{-3}$) and a low aerosol loading episode (LALD; PM$_{10}$: 180±68µgm$^{-3}$, PM$_{2.5}$: 67±18µgm$^{-3}$). Changes in columnar aerosol properties (MODIS AOD: 0.609-0.937) were found consistent with near surface particulates. A steady shift in fine mode fractions (FMF: 0.01-1.00) between different episodes revealed the existence of a variety of particulates in wider size domains. Satellite derived FMF were found in line with near surface particulate ratio (0.41-0.61). Variable particulate ionic fractions (19-29%) during different aerosol loading episodes were observed. Higher fractions of secondary inorganic aerosols during onset and peak winter (26-29%) highlighted the influence of biomass burning events at IGP along with locally emitted particles. The lagging winter episode however clearly depicted a sharp reduction in particulate ionic species (19-23%). The variability in aerosol characteristics at middle IGP clearly identified the intra-seasonal variations which is associated with their modifying source strength and regional meteorology.
Stratospheric intrusions investigated using high-resolution global simulations from the NASA GEOS-5 model.

Early Career Scientist

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Abstract:

The influence of stratospheric intrusions on surface ozone (O$_3$) concentrations is explored using a combination of observational datasets and modeling tools. In October 2015, the Environmental Protection Agency (EPA) revised the US air quality standard for O$_3$ from 75 ppbv to 70 ppbv. In many communities around the country, mitigations on O$_3$ precursors will be the driver to remain below the air quality standard. However, natural injection of high levels of O$_3$ from the stratosphere can cause air quality exceedences, especially for communities at high elevations in the springtime. Despite decades of research, the influence of stratospheric intrusions on air quality is still poorly understood. Using a multitude of observational datasets, including flight campaign data, ozonesondes and air quality monitoring surface sites, in combination with GEOS-5 modeling and assimilation products, the transport of O$_3$ from the stratosphere toward the surface is assessed. By focusing on recent reports of high O$_3$ at monitoring station sites likely associated with stratospheric intrusions, we aim to provide the public with tools which are available in near-real time to be able to more rapidly identify the impact of stratospheric air as opposed to anthropogenic sources.
New insights into VOC emissions and chemistry using high-resolution chemical-ionization time-of-flight mass spectrometry (H3O+ ToF-CIMS).

Early Career Scientist

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Abstract:

We have recently developed a new chemical-ionization time-of-flight mass spectrometer using H3O+ reagent ion chemistry (PTR-MS). The H3O+ ToF-CIMS has <1Hz time resolution, with VOC limits-of-detection in the 10s to 100s of ppt's for many atmospherically relevant species. The high mass resolution and time-of-flight operating principle allow simultaneous measurement of more than 1000 VOC ion masses. Here we illustrate significantly improved ability to detect and quantify complex VOC chemistry, using results from recent H3O+ ToF-CIMS field deployment, and experimental work with reagent ion chemistry.

The H3O+ ToF-CIMS was deployed on the NOAA WP-3 aircraft during the SONGNEX 2015 campaign, targeting emissions from oil and natural gas extraction field in the United States. These regions emit many novel and unexpected hydrocarbon species, and can also be influenced by emissions from urban areas, fires, forests, and agriculture. Using context from the full mass range measurement, isomerically-specific interpretation of H3O+ ToF-CIMS ion masses can be accomplished. Additionally, the high time resolution and many available masses allow the separation of emission sources and chemically distinct air masses within a basin. For example, application of PMF analysis to H3O+ ToF-CIMS data from flights over the Denver-Julesburg field in Colorado has enabled the separation of this region's complex, mixed emission sources. Additional field deployment on a mobile laboratory has provided further detailed compositional information.

In further development of this instrument, we have evaluated the use of NO+ reagent ion chemistry in the ToF-CIMS for measuring VOCs. NO+ CIMS may be an extremely useful technique for measuring aromatics, small aliphatics, semivolatile saturated alkanes, and carbonyl isomers.
6.180 Universal Spot Sampler: a new approach with unlimited possibilities for the chemical characterization of ambient aerosols.

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Abstract:

Here we present a new approach for the time-resolved collection of concentrated samples of ambient aerosol particles. The Universal Spot Sampler combines the simplicity of filter sampling with the completeness and automatization of real-time instruments. The system used a three-stage, laminar-flow water “moderated” condensation technique to collect airborne particles as concentrated dry spots (1-mm dia.) or liquid suspensions. Up to 33 uninterrupted, dry samples can be collected for periods of time ranging from minutes, to hours, to a day. Different collection matrices are available depending on the analysis conducted in the laboratory for chemical characterization. The system is field deployable and can run unattended for weeks. The Universal Spot Sampler has been validated for time-resolved characterization of ambient concentrations of polycyclic aromatic hydrocarbons, and sulfate and nitrate. Dry samples collected with the Universal Spot samples have also been analyzed by 2D-Thermodesorption gas chromatography (2D-TDGC-MS) and nano-Direct ElectroSpray Ionization (nanoDESI) for characterization of the organic composition of airborne particle samples, and by Laser Induced Breakdown spectroscopy (LIBS) for elemental analysis.

The Universal Spot Sampler is also capable of collecting soluble and insoluble constituents of particles as concentrated liquid suspensions (~400 µL) eliminating the extraction and pre-concentration steps required for the analysis of samples collected on filters. Liquid samples can be directly analyzed on-line by liquid chromatography to identify and quantify water soluble inorganics and organics, as well as bioaerosols. Anions, cations and carbohydrates have been easily measured by ion chromatography.

These are only few examples of the possibilities that the Universal Spot Sampler can provide to the atmospheric aerosol research community for better characterization of the chemical properties of ambient particulate matter, in a time-resolved manner.

Early Career Scientist

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Abstract:

Isoprene is the largest contributor to volatile organic compounds emissions from vegetation. Isoprene epoxydiols (IEPOX), which are oxidation products of isoprene via RO₂ +HO₂ pathway, can reactive uptake to acidic aqueous particles and produce secondary organic aerosol (i.e., denoted as isoprene-OA). Previous ambient measurements have shown a strong association between isoprene-OA and anthropogenic sulfate. However, due to the convoluted relationships between sulfate, particle acidity, and particle water, the mechanisms of the sulfate control on isoprene-OA formation is uncertain. In this study, we investigate the effects of sulfate on isoprene-OA formation through airborne measurements in the southeastern United States as part of the Southeast Nexus (SENEX) field campaign. We sampled downwind from two power plants (i.e., Harllee Branch and Scherer) in Georgia and found that isoprene-OA formation was only enhanced in Harllee Branch plume, but not in Scherer plume. This is because Harllee Branch emitted more sulfur dioxide (SO₂) than Scherer and more aerosol sulfate was produced downwind. The contrast between two power plants provides an opportunity to examine the magnitude and mechanisms of sulfate on isoprene-OA formation. We estimate that 1 µg sm⁻³ decrease in sulfate would decrease isoprene-OA by 0.23±0.08 µg sm⁻³. Further, we find that sulfate controls isoprene-OA formation in the power plant plume by enhancing both particle surface area and particle acidity, which enhances IEPOX uptake to particles and subsequent aqueous-phase reactions, respectively. Finally, we use these findings to explain the observed relationships between isoprene-OA, sulfate, particle acidity, and particle water in previous field studies.
6.185 New method for the calibration of an Aerosol Photo-Acoustic Spectrometer for in-situ measurement of Black and Brown Carbon.

Early Career Scientist

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Abstract:

Black carbon (BC) is estimated to have a strongly positive radiative effect on climate that is second only to CO₂ and methane. However, there are large uncertainties in the measurement of BC absorption, and the even greater uncertainty in quantifying the effect of organic coatings enhancing BC absorption and absorption from the organic coatings themselves (known as Brown Carbon). Moreover, while it is known that biomass burning (BB) accounts for a large fraction of black and brown carbon emissions, field studies directly measuring BB absorption are limited. The University of Wyoming has constructed an Aerosol Photo-Acoustic Spectrometer (PAS) based on the instrument developed by Lack et al. (2006) for the quantification of absorption in pertinent aerosols. This instrument has been deployed in a mobile laboratory to measure absorption properties of aerosols emitted during wildfires. The PAS has two cells at 405 nm and two cells at 660 nm wavelength which, coupled with a thermodenuder, measure absorption enhancement from organic coatings and brown carbon. The existing method for calibration of a multi-pass PAS is to use ozone. However, the absorption cross section of ozone at 405 nm is small enough to render direct calibration of this channel difficult without extremely large ozone concentrations. Here we present a new technique for calibration of the PAS, utilizing an Aerodyne Cavity Attenuated Phase Shift - Single Scattering Albedo (CAPS-SSA) instrument at both 660 nm and 405 nm. Absorbing aerosols (Aquadag, fullerene soot, or dried polystyrene latex) are size-selected using a Differential Mobility Analyzer (DMA) and passed through both the CAPS-SSA and PAS in series. From the CAPS-SSA measurement, both extinction and scattering will be quantified, and when subtracted, an absorption value. From this we reliably correlate our PAS acoustic response to absorption (Mm⁻¹) at each wavelength.
6.190 Characteristics of atmospheric depositions of ionic and carbonaceous components in North and South East Asia.

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Abstract:
Atmospheric deposition process is important to evaluate lifetimes and budget of atmospheric components. Deposition amounts of sulfur and nitrogen compounds have been evaluated not only in East Asian region but also worldwide. On the other hand, atmospheric deposition of carbonaceous components including organic carbon (OC), elementary carbon (EC) were monitored only at a few sites in Europe, North America and Africa, which will obscure removal process and atmospheric concentration distribution of those components. In this study, ionic and carbonaceous components in precipitation and aerosol are monitored at urban, rural and remote sites in Japan and Thailand, and the characteristics of atmospheric deposition amounts were evaluated.

Field observations have been implemented at Japanese stations since May 2011 and Thailand stations since September 2015. Wet deposition samples were collected by rain samplers, and dry deposition samples were collected by low volume aerosol samplers. Concentrations of Cl-, NO3-, SO42-, NH4+, Na+, K+, Mg2+, Ca2+ were measured by ion chromatography, EC and OC by the IMPROVE protocol. Wet deposition amounts were calculated as the products of aqueous concentration and precipitation amounts, and dry deposition amounts were as the products of air concentrations and deposition velocity estimated by the Inferential Method.

Atmospheric deposition amounts of carbonaceous components of OC and EC at Sado (remote) and Tokyo (Urban) sites from 2011 to 2012 were 2199.1 mg/m², 166.0 mg/m² and 2272.6 mg/m², 80.0 mg/m², respectively. OC was largely occupied in dry and wet deposition of carbonaceous components. Wet deposition of carbonaceous and sulfur components accounted 70%-90% of total deposition. Anthropogenic EC emission in Japan was comparable with EC atmospheric deposition. On the other hand, anthropogenic OC emission was much lower than OC deposition. This implies that biogenic OC emission and transportation of OC from outside of the region would be significant.
6.191 Formaldehyde and ethane variability in central Mexico from ground-based FTIR measurements.

Early Career Scientist

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Abstract:

The variability of formaldehyde (HCHO) and ethane (C₂H₆), two important organic molecules relevant to atmospheric chemistry involving photochemical reactions and direct emissions from biomass burning and combustion processes, were studied in central Mexico. Three years of data from ground-based solar absorption infrared measurements acquired at two stations: South of Mexico City at the UNAM campus (N19.32°, W99.17°, 2260 m.a.s.l.) and at the high-altitude Altzomoni NDACC station (N19.18°, W98.65, 3985 masl) were used to retrieve total vertical columns of these gases.

Results will be presented with time-series of these gases at two sites with close proximity (<60 km) but a large altitude difference (>1700 m), which in conjunction improve our current understanding of their vertical distribution, lifetimes and transport processes. These results will help in the near future for comparison studies with other ground-based techniques (MAX-DOAS), satellite-based measurements (TROPOMI, TEMPO, etc) and for improving the information fed and outputs in the global chemistry transport models.
A Low Power, High Mobility Cavity Ring-Down Spectroscopy Analyzer for In Situ Measurements of CO2, CH4, and H2O.

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Abstract:

Greenhouse gas accumulation has contributed to the changes in environments across the globe. Monitoring these fluctuations on global and local scales will allow scientists to better understand contributions that are made from biogenic and anthropogenic sources. This has led to the deployment of analytical instrumentation of all types to the most remote areas as well as the most densely populated areas. This however requires instruments to be precise, versatile, robust, and most importantly have power requirements that are as not limited by location, i.e. low enough power consumption to run off of batteries or even solar array.

Here we present a full greenhouse gas analyzer that utilizes a novel method of CRDS to measure carbon dioxide, methane and water vapor that consumes only 25W and still maintains long term stability to allow for averaging time of over 3 hours. Measurements have a 1-σ precision of 30 ppb for CO₂ and 300 ppt of CH₄ with 5 minutes of averaging; and with measurements of 3 hour averages reaching precisions down to 40 ppt of methane. Additionally this new flavor of CRDS has allowed for an overall increase in measurement dynamic range from traditional CW-CRDS measuring methane up to 1000 ppm and carbon dioxide up to several percent. We will present supplemental data acquired using this <11 kg analyzer, including soil respirations using closed static chambers and 10 m tower measurements from Santa Clara, CA.
6.193 ELEMENTAL ANALYSIS OF FINE ATMOSPHERIC AEROSOLS FROM A SITE IN MEXICO CITY.

Early Career Scientist

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Abstract:

Atmospheric aerosols in the Metropolitan Area of Mexico City (MAMC) still represent a major problem for air quality. Although the Atmospheric Monitoring System (also known as RAMA) measures the mass concentration of fine aerosols, it is necessary to obtain more information regarding the composition and the origin of these aerosols. It has been found that elemental analyses of aerosol samples, covering as many elements as possible, are a valuable method to identify several emitting sources.

As a consequence, a study was performed in the Winter of the year 2015 in a Southwestern site in the MAMC (Ciudad Universitaria), collecting PM$_{2.5}$ samples with a MiniVol sampler. This work is part of a wider study focused to fully characterize aerosols at this site. An X-ray Fluorescence (XRF) spectrometer (based on an Rh X-ray tube) built to analyze environmental samples [1], was used to analyze the sample set. A total of 16 elements (Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, and Pb) were detected in most samples. Mean concentrations are presented and compared to previous studies carried out in the MAMC [2].

The calculation of enrichment factors relative to Si were useful to find out which elements have a crustal or a human origin. Additionally, cluster analysis applied to the elemental concentration results in a possible identification of emitting sources. Moreover, possible relationships to organic and elemental carbon contents in the aerosols are suggested.

Work supported in part by DGAPA-UNAM (contract IN-102615) and the ACU project. The technical assistance of M.I. Saavedra is acknowledged.

6.194 Isotopic Methane and Ethane-to-Methane Ratio Analysis Using a Cavity Ring-Down Spectrometer.

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Abstract:

Natural gas analysis and methane specifically have become increasingly important by virtue of methane’s greenhouse warming potential compared. Large uncontrolled leaks, such as the recent one from southern California, originating from uncapped wells, storage facilities and coal mines have increased the total global contribution of methane missions even further. Determining the specific fingerprint of methane sources, by quantifying δ13C values and C2:C1 ratios, provides us with means to understand processes yielding methane and allows for sources of methane to be mapped and classified through these processes; i.e. biogenic or thermogenic, oil vs. gas vs. coal gas-related. In this study we present a fully developed Cavity Ring-Down Spectrometer (CRDS) that precisely measures 12CH4 concentration and its 13C H4 isotope concentration, yielding δ13C measurements, C2H6 concentration, along with CO2 and H2O. This provides realtime continuous measurements without an upfront separation requirement or multiple analyses to derive the origin of the gas samples. The highly sensitive analyzer allows for measurements of scarce molecules down to sub-ppb 1-σ precision in 5 minutes of measurement; with CH4 <0.1ppb, δ13C <1‰, C2H6 <1ppb and CO2 <1ppb. To complement this work, we provide the analysis of different methane sources providing a 2-dimensional mapping of methane sources as functions of δ13C and C2:C1 ratios; which can be thought of as a modified Bernard Plot. This dual ratio mapping can be used to discriminate between naturally occurring biogenic methane sources, naturally occurring enriched thermogenic sources, and natural gas distribution sources.

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Abstract:

Mediterranean-climate regions like California’s San Joaquin Valley are subject to severe wintertime particulate pollution affecting public health. We present maps of episodes and particulate diagnostics to aid diagnosis and amelioration. For abatement at sources, we require an understanding of sources and transport. Remote sensing should be of aid, but radiance-to-particle relationships are far different from methods which have been of use in the Eastern USA, Northern and Central Europe. Here are the problems: (a) Thin if very polluted mixed layers (MLs) yield optical depths, AOD, near the detection level, (b) bright and quite variegated surfaces (c) Unusual particle composition (e.g., predominance of NH4NO3 and fireplace burning aerosol), which complicate the relationship of AOD to PM2.5. Specialized analysis of MODIS-Aqua data to obtain AOD using the multi-angle (MAIAC) technique employed by Lyapustin and Wang. Meteorological analyses like NOAA’s Rapid Analysis Product (RAP, or newer products like HRRR), which are formulated to remain close to observations (e.g., of water), provide useful ML information corroborated by DISCOVER-AQ in-situ and lidar observations. The many PM2.5 measurements allow a calibration of these products and thus maps of aerosol on many successive aerosol buildups. These calibrations also allow insight into compositional information relevant to MODIS retrievals, the product of aerosol density and specific scattering. We have found that the rich in-situ, lidar, and sun-photometer data sets of NASA’S DISCOVER-AQ data set (2013) of great aid. We will highlight the most interesting of many intercomparisons possible with this rich data set. We conclude with a description of new work to extend these insights to similar regions, e.g. the Imperial Valley of California, the Po Valley and maritime Southern Europe, and the litoral regions of Israel.
Estimation of global methane fluxes using satellites, moving from GOSAT to TROPOMI.

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Abstract:
In the past years, there has been a large international scientific effort to develop instrumentation and data processing tools for estimating greenhouse gas emissions from satellite measurements. For methane these efforts concentrate currently on the use of data from the Japanese GOSAT instrument, which has been in orbit since 2009. Its value increases with each new year of data that is added to the record, covering most of the new phase of global methane increase, as well as important climatological disturbances in midlatitudes (e.g. the 2010 Moscow drought) and the Tropics (the 2010 Amazon drought, and ENSO). The interpretation of the data using models contributed strongly to the ongoing and sometimes heated discussions about the drivers of the renewed increase in global methane. We will discuss the main findings and limitations using GOSAT data. Meanwhile the community is preparing for the launch of Sentinel 5 precursor TROPOMI, which will complement GOSAT in the end of the year with a considerable improvement in measurement coverage. We will discuss the prospects of this new measurement capability for the quantification of methane emissions across a range of scales, and developments on the data processing side that will be needed to explore its full potential.
Abstract:

Chlorine can have significant impacts on several aspects of atmospheric chemistry, from the formation of tropospheric ozone to the oxidation of methane and non-methane hydrocarbons, and the cycling of nitrogen and sulphur. An accurate assessment of the role played by chlorine in the chemical processes of the lower atmosphere is complicated by an incomplete understanding of its sources, sinks and distribution. We present a set of observations of inorganic chlorine (Cl\(_2\), ClNO\(_2\), particulate chloride) and supporting data (N\(_2\)O\(_5\), O\(_3\), NO\(_x\), VOC, photolysis rates and meteorological parameters). This dataset provides insight into the geographical and seasonal variability of chlorine in Northern Europe. The measurements were taken between 2014 and 2016 at different locations in Britain: an urban site 200 km from the sea (Leicester), a coastal site in the south-west, experiencing clean air from the Atlantic Ocean (Penlee Point, Cornwall) and a coastal site in the east experiencing clean air from the North Sea or polluted air from inland (Weybourne, Norfolk). ClNO\(_2\) was almost always present at night with peak concentrations of the order of hundreds parts-per-trillion; Cl\(_2\) was observed only occasionally, with concentrations of a few tens of parts-per-trillion. Distinct seasonal patterns were observed for ClNO\(_2\) with springtime maxima and summertime minima. The implications of the ubiquitous presence of these forms of inorganic chlorine for the
production of ozone and other pollutants in Northern Europe are investigated and discussed with the help of the NAME atmospheric dispersion model and of the GEOS-Chem 3-D chemical transport model.
A sun photometer to connect local and remote sensing of aerosols.

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Abstract:

Aerosol extinction is a fundamental parameter for visibility and for the direct forcing of climate visibility. Vertical profiles are an essential complement to satellite measurements of column optical depth. A miniature, four-wavelength sun photometer has been developed for vertical profiles of aerosol extinction. It weighs about 360 g, is low power, and uses inexpensive components. It is suitable for routine profiles on small balloons and UAVs. Flight data show that it can distinguish aerosol optical depths of about 0.01 and measure the scattering phase function even with very low aerosol optical depths.
Tracing sources of airborne particulate matter using stable lead and strontium isotopes in Kyrgyzstan.

Early Career Scientist

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Abstract:
Central Asia is dominated by an arid climate and desert-like conditions, leading to the potential of long-range transport of desert dust. One potential source of dust to Central Asia is the Aral Sea, the surface area of which has receded in size from 68,000 km$^2$ to 8,444 km$^2$, largely as the result of water diversion. We employed Sr and Pb stable isotope ratios, along with detailed elemental composition, to explore the contribution of long-range transport of Aral Sea sediments, as well as other potential sources of dust to Central Asia. Ambient PM$_{10}$ samples from two sites (Bishkek and LIDAR) in Kyrgyzstan both during dust and non-dust events, along with resuspended sediments, and local soils were collected. The average $^{87}$Sr/$^{86}$Sr ratio of the Aral Sea sediments was found to be 0.70992 (0.70951-0.71064). In contrast, the local soils in Kyrgyzstan exhibit an average $^{87}$Sr/$^{86}$Sr ratio of 0.71579 (0.71448-0.71739), which is significantly different than the Aral Sea sediments. The PM$_{10}$ collected in Kyrgyzstan have an average $^{87}$Sr/$^{86}$Sr ratio of 0.71177 (0.70946-0.71335), indicating a complex mixture of contributions, which may include long-range transport of Aral Sea sediments, dusts from regional deserts, and local soils. Similar Sr isotope ratios were observed during the dust and non-dust events at both sites, indicating that the Aral Sea sediments only have a minimal effect to air quality in Kyrgyzstan. Elemental analysis and Pb isotope ratios were helpful in refining the source regions. Stable Pb isotope ratios confirm that, although the Aral Sea region may be a minor source of aerosol Pb in Kyrgyzstan, there is at least one other source that was not measured in this study. While the isotope and elemental data both indicate an anthropogenic source, long-range dust transport from Africa or the Middle East cannot be ruled out as sources of PM$_{10}$ to Central Asia.
6.205 Indirect Evidence on the Composition and Origin of Ultrafine Atmospheric Particles in the High Arctic.

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Abstract:

Long-term observations show that nanoparticle formation, i.e., nucleation events are common in the summer-time high Arctic\(^1,2\), and are linked to local photochemical activity. However, theories disagree on the chemical precursors of resulting nanoparticles and on the species involved in their subsequent growth, thereby making it difficult to predict their impact on climate. Inorganic compounds can be produced by oxidation of sulfur dioxide emitted mostly from lower latitudes\(^3\) or marine organosulfur species\(^4\), whereas organic compounds can be produced from marine condensable species\(^2\) or microgel fragments\(^5\). To identify which species are involved in nanoparticle growth in the high Arctic and thus better characterize their composition, here we report measurements of their size distribution and for the first time the volatility of monodisperse particles having diameters ≤200 nm. The volatility measurements suggest that organic compounds are not present on nanoparticles to any large extent. The air mass origin identified for the events observed in this study, by means of back trajectories, is located over the central Polar Region where no anthropogenic influence is expected. Along with gas concentration and meteorological data, our observations provide strong evidence that a predominant fraction of the 12-nm particle population is ammoniated sulfates.

References:

6.206 Source of SO2 and fine sulfate particles (<0.45 µm) in the Arctic summer.

Early Career Scientist

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Abstract:
Aerosols play an important role in the Arctic climate change, and despite their importance, crucial gaps in our understanding/estimation of their effects and sources remain. Size-segregated aerosol particles were collected on board the Canadian Coast Guard Ship (CCGS) Amundsen in the Arctic during July 2014, to quantify the sulfate aerosol concentration and measure the sulfur isotopic composition to address the contribution of sea salt/non-sea salt and anthropogenic/biogenic sources to the growth of aerosol size fractions in the Arctic atmosphere. Results show that sea salt sulfate aerosols were especially high in coarse mode aerosols, however, more than 97% of fine aerosols (<0.45 µm) were from non-sea salt sources, and more than 63% of aerosols less than 0.49 microns in diameter (fine aerosol) and 86% of SO2 were from biogenic sources. A comparison of the isotope values (δ34S) for SO2 and fine aerosols suggests that they originated from the same sources (except for two samples) and gas-to-particle conversion occurred during most sampling periods.
6.208 16 years of carbon monoxide observations from MOPITT.

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Abstract:

Measurements Of Pollution In The Troposphere (MOPITT) on the NASA Terra spacecraft has been measuring the global atmospheric abundance of carbon monoxide (CO) since March 2000. CO is mainly produced by incomplete combustion from both natural fires and anthropogenic activities and is also a product of chemical reactions with other air pollutants. CO plays an important role in atmospheric chemistry and climate because it is a dominant sink for the hydroxyl radical (OH) and thus affects the abundance of greenhouse gases methane (CH$_4$) and ozone (O$_3$). Since CO is a pre-cursor to greenhouse gases, anthropogenic emissions of CO have a small but significant indirect radiative forcing of 0.22 W/m$^2$. Satellite measurements of carbon monoxide are used to understand how pollution is emitted and transported globally, from large scale fires to urban sources. I will present an overview of the MOPITT mission and show recent science results using MOPITT CO data, including highlights on how MOPITT data are assimilated for understanding model chemistry and emissions, global trends in CO concentrations and the impact of the 2015 ENSO on fires in Indonesia.
6.209 First Lidar measurements in the Brazilian Northeast to study transatlantic transport of Saharan aerosols.

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Abstract:

The Lidar technique has been widely used in atmospheric sciences in recent years because it provides effective information about atmospheric aerosol optical properties and its evolution. The presence of Saharan aerosol particles in the Brazilian Amazon region has long been observed during intensive measurements of Lidar field campaigns, however long term measurements are required to improve and complement the satellite observations, by providing highly time and altitude resolved data. The Laser Environmental Application Laboratory (LEAL) at the Nuclear and Energy Research Institute (IPEN, São Paulo, Brazil) has developed a depolarization lidar system which was recently installed at the Department of Atmospheric and Climate Sciences of the Federal University of Rio Grande do Norte (UFRN) in the city of Natal in the Brazilian Northeast (5°50’29 S, 35°11’57 W, s.l.). This new Latin American Lidar station has started operation in early February 2016 to provide routinely vertical profiles with high spatial and temporal
resolution of aerosol optical properties mainly for identifying aerosol layers originating in the African desert. This work presents the first results retrieved from these Lidar observations between February and August 2016, focused into time series evolution of aerosol backscattering coefficients during the period of measurements and including the identification of aerosol layers with potential dust particles by depolarization sensitive backscatter profiles.

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Abstract:
The Wideband Integrated Bioaerosol Spectrometer (WIBS) measures the equivalent optical diameter (EOD) and the fluorescence of individual particles in three spectra bands when the particle is excited at two wavelengths: 280 nm and 370 nm. Here we describe measurements made using the WIBS during an 18-day period from May 20-June 6, 2015 in the city of Leon, Spain. Hourly filter samples were also taken for pollen count analysis. A time series of wind speed, relative humidity (RH) and the fluorescence fraction (FF), defined as the ratio of all fluorescing particles to the total particles detected, show a clear diurnal cycle of all three parameters but out of phase with one another. The maxima in FF always occurs near midnight following the decrease in wind speed to near zero. The FF and RH are increasing at the same time but the FF maximizes before the RH. The cross correlational analysis shows that the FF reaches its maximum approximately two hours after the decrease in wind speed and three hours before the maximum in RH. Fluorescing particles are classified into seven types based on previous analysis and compared with a library of bacteria, fungi and pollen. Those particles that don't match library types are classified as “other”. An analysis of the bioaerosol components shows that the increase in FF is partially due to the decrease in the total particle concentration but that the total bioaerosol concentration increases by about 60%, dominated by the increase in pollen-like and “other” bioaerosols. The bacteria remains constant throughout the day indicating that it is not dependent on wind speed or RH. We also observe that bacteria, fungi and pollen are 16%, 20% and 25% of the total bioaerosol population, respectively. The other remaining 37% of the bioaerosols remain to be identified.
An investigation on the origin of regional spring time ozone episodes in the Western Mediterranean and Central Europe.

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Abstract:

For the identification of regional spring time ozone episodes, rural EMEP ozone measurements from countries surrounding the Western Mediterranean (Spain, France, Switzerland, Italy, Malta) have been examined with emphasis on periods of high ozone, according to the daily variation of the afternoon (12:00 – 18:00) ozone. For two selected high ozone episodes in April-May 2008, composite NCEP/NCAR reanalysis maps of various meteorological parameters and/or their anomalies (geopotential height, specific humidity, vertical velocity omega, vector wind speed and temperature) at various tropospheric pressure levels have been examined together with the corresponding satellite IASI ozone measurements (at 3 and 10 km), CHIMERE simulations, vertical ozone soundings and HYSPLIT back trajectories.

The results show that high surface ozone is measured at several countries simultaneously over several days. Also, the examined spring ozone episodes in Western Mediterranean and Central Europe are linked to synoptic meteorological conditions very similar to those recently observed in summertime ozone episodes over the Eastern Mediterranean (Kalabokas et al., ACP, 2013; Doche et al., ACP, 2014; Kalabokas et al., TellusB, 2015), where the transport of tropospheric ozone-rich air masses through atmospheric subsidence influences significantly the boundary layer and surface ozone concentrations.
In particular, the geographic areas with observed tropospheric subsidence seem to be the transition regions between high pressure and low pressure systems. During surface ozone episodes, strong gradients of geopotential height and temperature are observed, together with high positive omega vertical velocity (downward transport) and low specific humidity (dry conditions), at all examined pressure levels below 500 hPa. IASI satellite measurements show extended areas of high tropospheric ozone over the adjacent to the anticyclones low pressure systems, which influence significantly the boundary layer and surface ozone concentrations within the anticyclones by subsidence and advection, in addition to the photochemically produced ozone, resulting to exceedances of the 60 ppb standard.
Abstract:

The Copernicus Atmosphere Monitoring Service (CAMS) delivers several operational services to address environmental concerns such as the air quality at the global and European scales, the health of ecosystems, the levels of stratospheric ozone, and the distribution of dust, sand, smoke and volcanic aerosols. CAMS delivers a large set of products that support policymakers, business, scientists and citizens, and that provide enhanced atmospheric environmental information.

The poster discusses the information generated by the CAMS system including daily near-real-time analyses and forecasts of global atmospheric composition. It also provides consistent multi-annual global datasets of atmospheric composition from reanalyses with a model/assimilation system. Daily near-real-time European multi-model air quality analyses and forecasts, reanalyses providing consistent annual datasets of European air quality and other policy-relevant products are also made available. In addition, CAMS provides solar and UV radiation fields that support the planning, monitoring, and efficiency improvements of solar energy production. Quantitative information on UV irradiance for downstream support applications related to health and ecosystems. Greenhouse gas surface flux inversions for CO\textsubscript{2}, CH\textsubscript{4} and N\textsubscript{2}O allow the monitoring of their evolution in time. Climate forcing from aerosols, long-lived (CO\textsubscript{2}, CH\textsubscript{4}) and shorter-lived (stratospheric and tropospheric ozone) compounds is also derived.
6.213 More than 15 years long-term monitoring of hydroxyl radicals at the GAW station Hohenpeissenberg.

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Abstract:

Hydroxyl radicals (OH) are known to be the major daytime detergent of the atmosphere playing a key role in the initiation of oxidation processes and in tropospheric ozone and particle formation. Their abundance is governed by the presence of water vapour, ozone, sun light, as well as by biogenic and anthropogenic pollutants. Anthropogenic emissions, change in land use and climate change have being altering the atmospheric composition constantly with potential impact on the atmospheric self-cleaning capacity. Whilst several field campaigns have measured OH radicals for short term studies, hardly any continuous long term observations are available. So far, the long term behaviour of global OH concentrations has been solely estimated indirectly by the decay of certain tracers with well-known emissions, such as methylchloroform.

Here, we report about long term OH measurements, performed since 1998 on an almost continuous base at the global GAW station Hohenpeissenberg, South Germany. OH is being measured alongside with sulfuric acid, other trace gases and meteorological parameters routinely, using chemical ionization mass spectrometry (CIMS). Diurnal and seasonal cycles, as well as the time series are presented and the possible impact of change in the atmospheric composition over almost eighteen years is investigated.
6.214 Subseasonal variability of surface ozone in the Mexico City basin.

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Abstract:

Concentrations of winter and summer ozone in the Mexico City basin were investigated for variability by phase of the Madden-Julian Oscillation (MJO). The MJO is the leading mode of atmospheric variability on the subseasonal (30-60 day) time scale, and changes in large-scale tropospheric circulation occur in both the tropics and extratropics in response to the convective heating anomalies of the MJO. The primary hypothesis tested in this study was whether large-scale changes in tropospheric circulation would affect surface ozone concentrations in Mexico City. Mexico City's ozone, with nearly 30 years of hourly measurements at stations around the metropolitan area, offer a unique chance to investigate both local and teleconnected influence of the MJO because, at 19 degrees north latitude, Mexico City experiences both tropical (in summer) and extratropical (in winter) synoptic-scale circulation patterns. The primary findings of the study were: (1) surface ozone concentrations did exhibit statistically significant (p < 0.05) variability by phase of the MJO in both summer and winter. In summer, highest ozone concentrations occurred on days when the MJO was active and in phase 6, while lowest concentrations occurred on days when the MJO was active and in phase 1. In winter, highest ozone concentrations were occurred on days when the MJO was active and in phase 2, while lowest concentrations occurred on days when the MJO was active and in phase 8. (2) Anomalies in both total cloud cover and surface UV-B radiation were found that strongly supported this variability. (3) Finally, in both summer and winter, days with above-normal ozone featured anomalies in upper-tropospheric circulation supporting reduced cloud cover, and days with below-normal ozone featured anomalies in upper-tropospheric circulation supporting enhanced cloud cover.
Constraining regional and national fossil fuel CO\textsubscript{2} emissions using atmospheric observations of CO\textsubscript{2} and 14CO\textsubscript{2}.

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Abstract:

Recent US policy measures and international commitments, such as the President’s Climate Action Plan and the US “Intended Nationally Determined Commitment” (INDC) to the UNFCCC’s COP21, call for a reduction in US greenhouse gas (GHG) emissions over the next several decades. For example, the US INDC aims at a reduction of the annual net CO\textsubscript{2} equivalent GHG emission by 25-28% with respect to 2005 levels by 2025. This raises the question of whether the US and other nations have the tools needed in order to detect and verify proposed emission reductions. Here we use the fact that $\Delta^{14}$CO\textsubscript{2} is a largely unbiased tracer of recently-added fossil fuel CO\textsubscript{2} in the atmosphere to construct an atmospheric inverse modelling framework to independently estimate fossil fuel CO\textsubscript{2} emissions from observations of CO\textsubscript{2} and $\Delta^{14}$CO\textsubscript{2}. We evaluate present and realistic future observing scenarios in an Observation System Simulation Experiment (OSSE), including the low end National Research Council recommendation of 5,000 $\Delta^{14}$CO\textsubscript{2} measurements per year. We show that given the present day observing scenario, this framework can estimate the US annual total emission to within 5%. Given an augmented yet realistic future observing scenario of 5,000 measurements per year over North America, we can estimate monthly total emissions over the US and several highly emissive regions (such as the Eastern US) with the same accuracy.
6.216 Tropical sources and sinks of carbonyl sulfide observed from space.

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Abstract:

Regional and global analysis of atmospheric carbonyl sulfide (COS) can provide a novel constraint on the carbon cycle (Campbell et al., Science 2008). However, observations of COS are sparse in tropical regions. Here we use the comprehensive data set of spaceborne measurements of the Michelson Interferometer for Passive Atmospheric Sounding to analyze its global distribution (Glatthor et al., GRL, 2015). Two major features are observed in the tropical upper troposphere around 250 hPa: enhanced amounts over the western Pacific and the Maritime Continent, peaking around 550 parts per trillion by volume (pptv) in boreal summer, and a seasonally varying depletion of COS extending from tropical South America to Africa. The large-scale COS depletion, which in austral summer amounts up to −40 pptv as compared to the rest of the respective latitude band, has not been observed before and reveals the seasonality of COS uptake through tropical vegetation. The observations can only be reproduced by global models, when a large vegetation uptake and a corresponding increase in oceanic emissions as proposed in several recent publications are assumed.