Aerosols, Health and Climate: Gigacity and Future

782. WE-Heraeus-Seminar

20 – 24 March 2023

at the Physikzentrum Bad Honnef, Germany

WILHELM UND ELSE
HERAEUS-STIFTUNG

Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the 782. WE-Heraeus-Seminar:

Atmospheric aerosols are strongly affecting human health and climate in the Anthropocene. Airborne transmission by aerosols and droplets is important for the spread of viruses, including the severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2), and fine aerosol particles (e.g., PM2.5) are associated with several adverse health effects causing significance increase of premature death and mortality. Besides, aerosols can also interact with radiation and clouds, regulating the earth energy budget, water cycle and the climate.

Despite its importance, the large knowledge gap in aerosol formation and interaction processes introduce large uncertainties, preventing us from developing effective measures to mitigate adverse impact of aerosols. This seminar will deal with these questions in depth, especially the most challenging ones and key bottlenecks that can be resolved by an interdisciplinary approach through collaborations between experimentalists and modeler, and between researchers from different disciplines. We will place the discussion of aerosol impact in this context of gigacities, typical examples of the huge human influence on the earth, aiming at bringing enlightenment to the future development of human society.

The seminar will focus on the following topics:
- Atmospheric aerosols and air pollution
- Aerosol, health and pandemic
- Aerosol and climate
- Interdisciplinary emerging techniques and knowledge

Potential participants are both excellent established scientists who, as invited speakers, can give an up-to-date overview of the various aspects of aerosol research, and outstanding young scientists and doctoral students who are eager to think outside the box and would like to learn an interdisciplinary approach to break the borders of different areas.

Scientific Organizers:

Prof. Dr. Yafang Cheng — MPI for Chemistry, Mainz, Germany
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Prof. Dr. Markku Kulmala — University of Helsinki, Finland
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Prof. Dr. Hang Su — MPI for Chemistry, Mainz, Germany
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Introduction

Administrative Organization:

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Registration:

Elisabeth Nowotka (WE Heraeus Foundation) at the Physikzentrum, reception office Monday (17:00 h – 21:00 h) and Tuesday morning

Program

Program

Monday, 20 March 2023

17:00 – 21:00	Registration	
18:00	*BUFFET SUPPER and informal get-together*	

Tuesday, 21 March 2023

08:00	*BREAKFAST*	
09:00	Scientific organizers	**Welcome words**
09:10 – 09:55	Tuukka Petäjä	**Strong control of aerosol-cloud interactions by emissions from the boreal forests**
09:55 – 10:30	Renyi Zhang	**Formation and human health impacts of ultrafine particles**
10:30 – 11:00	*COFFEE BREAK*	
11:00 – 11:35	Mischa Bonn	**Protons at the water surface: surface pH versus bulk pH**
11:35 – 12:10	Colette Heald	**Natural aerosol impacts on air quality and climate**
12:10 – 12:45	Zhanqing Li	**Air quality, climate changes and their intertwined relationships in Asia**
12:45	*LUNCH*	

Program

Tuesday, 21 March 2023

13:40 – 14:25	Jos Lelieveld	**Global impacts of particulate air pollution on public health**
14:25 – 15:00	Gregory Carmichael	**Multi-sensor exposure estimates to help better understand air pollution impacts**
15:00 – 15:35	Christian Pfrang	**Research into aerosols, films and wider atmospheric chemistry**
15:35 – 16:05	*COFFEE BREAK*	
16:05 – 16:40	Jasper Kirkby	**Perspective on atmospheric particle formation from the CERN CLOUD experiment**
16:40 – 17:15	Heikki Junninen	**Particle filtration efficiency and breathability of commonly used facial masks**
18:00	*DINNER*	

Program

Wednesday, 22 March 2023

08:00	*BREAKFAST*	
09:00 – 09:35	Ulrike Lohmann	**Aerosol impact on climate by impacting warm and mixed-phase clouds**
09:35 – 10:10	Ulrich Pöschl	**Physical, chemical, and biological multiphase processes influencing climate and health in the Anthropocene**
10:10	**Conference photo** *(in the front of the lecture hall)*	
10:10 – 10:40	*COFFEE BREAK*	
10:40 – 11:15	Doug Worsnop	**Atmospheric aerosol chemistry: From the boreal forest to gigacities**
11:15 – 11:50	André Prevot	**Recent findings on air pollution sources, processes and links to health in Indian and Chinese cities**
11:50 – 12:25	Veli-Matti Kerminen	**Current understanding on atmospheric new particle formation**
12:25	*LUNCH*	

Program

Wednesday, 22 March 2023

13:40 – 14:15	Thomas Münzel	**Effects of air pollution on cardiovascular disease**
14:15 – 14:50	Matei Georgescu	**The twin forcing agents of urban climates: impacts and perspectives on future directions**
14:50 – 15:50	**Poster madness**	
15:50	**Poster** *& COFFEE*	
18:30	*DINNER*	
19:30	**Poster**	

Program

Thursday, 23 March 2023

08:00	*BREAKFAST*	
09:00 – 09:45	Meinrat Andreae	**Aerosols: A deadly climate mitigation**
09:45 – 10:20	Neil Donahue	**Peroxy radicals really are at the center of the universe**
10:20 – 10:50	*COFFEE BREAK*	
10:50 – 11:25	Ilona Riipinen	**Insights into aerosol-cloud-climate interactions in environments with limited human influence**
11:25 – 12:00	Merete Bilde	**Plastic in the air: results from laboratory experiments on bubble mediated aerosol formation**
12:00 – 12:35	James Allan	**The continuing evolution of combustion aerosols in the UK**
12:35	*LUNCH*	
13:40 – 14:15	Thorsten Hoffmann	**Chemistry in nanometer particles: Laboratory studies on particle size-dependent aerosol chemistry**
14:15 – 14:50	Eberhard Bodenschatz	**Human aerosols and drops and the associated transmission risk of respiratory pathogens**
15:20	**Excursion** *& COFFEE*	
19:00	*HERAEUS DINNER* *(social event with cold & warm buffet with complimentary drinks)*	

Program

Friday, 24 March 2023

08:00	*BREAKFAST*	
09:00 – 09:35	Birgit Wehner	**Vertical profiling of aerosol particles in Arctic regions**
09:35 – 10:10	Tom Kokkonen	**Interactions of boundary layer dynamics, urban climate, and air quality in the Chinese Gigacity**
10:10 – 10:30	*COFFEE BREAK*	
10:30 – 11:30	**Panel discussion**	
11:30 – 12:00	Scientific organizers	**Rapporteur summary**
12:00 – 12:20	Scientific organizers	**Closing remarks & poster awards**
12:30	*LUNCH*	

End of the seminar and departure

NO DINNER for participants leaving on Saturday; however, a self-service breakfast will be provided on Saturday morning

Posters

Posters

Ajit Ahlawat	**Exploring low-level haze formation mechanisms using customized drone setup at Delhi**
Honey Dawn Alas	**Aerosols and the plight of the exposed: understanding personal exposure through microscale air pollution research**
Gianluca Armeli	**Machine learning-based prediction of the glass transition temperature**
Christof Beer	**A global climatology of ice-nucleating particles under cirrus conditions derived from model simulations with MADE3 in EMAC**
Thomas Berkemeier	**KM-SUB-LUNG – A multiphase kinetic model untangling the chemical and biological contributions of air pollution to oxidative stress in the lungs**
Federico Bianchi	**New-particle formation in polluted environment: A comparison between Beijing and the Po Valley**
Nadine Borduas-Dedekind	**Cool cloud chemistry for climate: Organic aerosol's ability to nucleate ice in mixed-phase clouds**
Jing Cai	**Long-term reduction of fine particulate matters in Beijing after the COVID lockdown: an investigation based on more than 3-year observations**
Liangduo Chen	**High number concentration of atmospheric sub-3 nm particles in polluted environment of East China: three years' observation at SORPES station**
Meng Gao	**Reduced aerosol pollution and intensified summertime rainfall under COVID-19 lockdown in India**

Posters

Florence Gregson	**Biomass burning organic aerosol: phase separation and viscosity**
Yufang Hao	**Molecular characterization of organic aerosols in two heavily polluted cities of India: seasonality and sources**
Tareq Hussein	**The frequency of sand and dust storms in the eastern mediterranean region and its possible health risk**
Hyun Gu Kang	**Oxidation products and aerosol yields from D5 siloxane, an emerging urban pollutant**
Peeyush Khare	**Elucidating variations in molecular-level speciation and sources of ambient organic aerosol in two megacities of North and South India**
Matthias Kohl	**Numerical simulation and evaluation of global ultrafine particle concentrations**
Matteo Krüger	**The kinetic laboratory compass: Using kinetic models and machine learning for experiment design**
Jingmin Li	**Investigation of the development of global aerosol regimes from pre-industrial times to the future based on global aerosol simulations and machine learning techniques**
Yuanyuan Li	**Enigma of biogenic VOCs in highland ecosystem: characteristics, emissions and sources based on in-situ observations at the Qinghai-Tibet Plateau**
Chaoqun Ma	**Importance of bias correction in data assimilation of multiple observations over Eastern China using WRF-Chem/DART**

Posters

Nan Ma	**A new airborne aerosol sampling system: development, validation, and application in vertical profile measurements of aerosol properties**
Fabian Mahrt	**Number of phases in organic aerosol mixtures is driven by difference in oxygen-to-carbon ratio of components**
Wenjun Meng	**Source contributions and drivers of physiological and psychophysical co-benefits from major air pollution control actions in North China**
Adam Milsom	**MultilayerPy: a tool for creating and optimising multi-layer models of aerosol and film processes**
Ashmi Mishra	**The effect of biological and atmospheric oxidants on proteins in the epithelial lining fluid**
Ruijing Ni	**Effects of long-term ambient fine particulate matter exposure on asthma: Evidence for children and adults**
Andreas Paul	**EURO 6 compliant gasoline vehicle emissions and SOA formation after aging in the photochemical emission aging flow tube reactor during steady state driving**
Jan-Hendrik Peters	**Glyoxal as a potential source of highly viscous aerosol particles**
Lu Qi	**The influence of different types of SOA on climate and human health**
Wiebke Rautenberg	**Heterogeneous chemistry and nanometer particle growth: laboratory studies on transesterification in nanometer-sized aerosol particles**

Posters

Anu-Maija Sundström	**Satellite proxies for estimating spatial variation of new particle formation**
Baptiste Testa	**Cirrus temperature ice nucleation abilities of soot from commercial aircraft engines**
Qiaoqiao Wang	**The export of African dust across the Atlantic and its impact over the Amazon Basin**
Yu Wang	**By how much can co-condensation of semi-volatile compounds alter clouds?**
Liwen Yang	**Budget analysis of gas-phase sulfuric acid concentration and its contribution to sulfate in polluted Yangtze River Delta, East China**
Guangjie Zheng	**The role of CO_2, ammonia, and organic acids in buffering atmospheric acidity: the distinct contribution in clouds and aerosols**

Abstracts of Talks

(in alphabetical order)

The continuing evolution of combustion aerosols in the UK

J.D. Allan[1,2], I. S. Wong[1], N. Watson[1], S. Alzahrani[1], Z. Bibi[1], Z. Chen[1], H. Coe[1], M. Flynn[1], D. Hu[1], O Oghama[1], T Bannan[1], A Voliotis[1,2], G. McFiggans[1], A. Lea-Langton[3]

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[3]Dept. of Mechanical, Aerospace and Civil Engineering, The University of Manchester, Manchester, UK

Combustion has traditionally been one of the greatest sources of particulate pollution in the UK. Previous assessments by the University of Manchester had shown that both diesel vehicles and domestic wood combustion are major contributors to primary aerosols in London and Manchester[1,2], however over the past decade, emissions have changed drastically. Vehicle particulate emissions in cities have reduced in line with the introduction of mandatory diesel particle filters and clean air zones, whereas the practice of burning wood has only increased in popularity. While the government has attempted to mitigate the effect of wood burning by mandating newer 'Ecodesign' stoves and banning the sale of wet wood, it is still thought to be one of the major contributors to ambient $PM_{2.5}$. And yet the representation of wood burning in inventories is highly uncertain.

Here we present recent work to characterize wood burning emissions, both in ambient and in the laboratory, to determine the magnitude and nature of wood burning emissions. This includes measurements at the Manchester Air Quality Supersite, mobile measurements using portable devices, fluxes using the eddy covariance technique applied to the Single Particle Soot Photometer (SP2) and analysis of datasets using machine learning to extract long term trends. Laboratory work has focused on fundamental combustion processes and the emissions from the latest designs of stoves. These ongoing studies are being used to further fundamental understanding of emissions and how these might affect human health, and upcoming experimental programmes will be discussed.

References

[1] Allan et al., Atmos. Chem. Phys. **10**, 647-668 (2010)
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Aerosols: A Deadly Climate Mitigation

MEINRAT O. ANDREAE
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Abstract

Atmospheric aerosols have counteracted the warming effects of greenhouse gases over the past century. This has provided some “climate mitigation”, but at great cost to human health. The cooling effect of aerosols has also prevented the true magnitude greenhouse gas warming from becoming evident. Over the coming decades, the role of aerosols in opposing global warming will wane, since their lifetime is short, and because there are powerful policy reasons to reduce their emissions. They will, on the other hand, continue to play a role in regional climate change, especially with regard to the water cycle. The health effects of aerosols, combined with the potentially very high sensitivity of the climate system, make sharp and prompt reductions in both aerosol and greenhouse gas emissions, especially CO_2, very urgent.

Atmospheric Aerosols versus Greenhouse Gases in the Climate of the 21st Century

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Extended Abstract

Aerosols have a wide range of climate effects, which can be broadly classified into direct (i.e., those based on the interaction of radiation with the particles themselves) and indirect effects (i.e., those that result from aerosols modifying the abundance and properties of clouds) (*1*). Aerosols reflect solar radiation back to space, thus reducing the heat absorbed by the Earth and thereby lowering its temperature. On the other hand, some aerosols (e.g., soot particles) absorb sunlight, thus warming the atmosphere, while still cooling the surface. This atmospheric warming suppresses convection, cloudiness, and precipitation. Shading of the surface by aerosols also reduces evaporation, which leads to less rainfall.

Cloud droplets form when supersaturated water vapor condenses on aerosol particles, so-called cloud condensation nuclei (CCN). In a given cloud, the more such particles there are, the more droplets will form. This makes the cloud more reflective, and therefore has a cooling effect on the planet. With more or less the same amount of water available in the cloud, however, the droplets in the more aerosol-rich cloud must be smaller. This makes it more difficult for them to grow into rain drops, and the clouds and water vapor persist longer in the atmosphere. Cloud convection dynamics is also affected, which in turn perturbs the global circulation (*2-4*).

Atmospheric aerosols have counteracted the warming effects of greenhouse gases (GHG) over the past century. This has provided some “climate protection”, but also prevented the true magnitude of the problem from becoming evident. Over the coming century, the role of aerosols in opposing global warming will wane, since their lifetime is short, and because there are powerful policy reasons to reduce their emissions. In contrast to the GHG, aerosol pollution has severe health impacts that affect most strongly the populations in the region where emissions take place. This has created strong incentives to reduce emissions by regulation and technological means, which has resulted in decreasing aerosol levels in most developed countries (*5*). Analysis of present trend and future scenarios leads to the expectation of worldwide decreases in the atmospheric aerosol burden over the 21st century, after a maximum in the early part of the century.

This will lead to a reduction of both the relative and the absolute magnitude of global aerosol climate effects, and the true impact of GHG increases on climate will be revealed. Aerosols will, on the other hand, continue to play a role in regional climate change, especially with regard to the water cycle. The can reduce rainfall in sensitive areas, and shift rainfall patterns towards more severe storm events.

Because of the reduction of climate forcing by aerosols in the 20th century, it is likely that the sensitivity of the climate system to GHG forcing has been underestimated (*6*). The end of significant “climate protection” by atmospheric aerosols, combined with the potentially very high sensitivity of the climate system, make sharp and prompt reductions in greenhouse gas emissions, especially

CO_2, very urgent. That such reductions are technically feasible and economically advantageous has been demonstrated recently (*7, 8*).

References and notes

1. M. O. Andreae, in *World Survey of Climatology. Vol. 16: Future Climates of the World* A. Henderson-Sellers, Ed. (Elsevier, Amsterdam, 1995) pp. 341-392.
2. M. O. Andreae *et al.*, *Science* **303**, 1337 (2004).
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8. N. Stern, *The Economics of Climate Change - The Stern Review* (Cambridge University Press, 2006), pp.

Plastic in the air: results from laboratory experiments on bubble mediated aerosol formation

Eva R. Kjærgaard[1], Freja Hasager[1], Andreas Massling[2], Sarah S. Petters[1], Marianne Glasius[1], M. Bilde[1]

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Plastic pollution is increasing in several components of the Earth system[1] in particular soil[2] and water.[3, 4] Recently it has become clear that plastic pollution is also transferred into the air with tire wear, industrial and agricultural activities,[9] and by bubble-mediated aerosolization of nano- and microplastics from surface ocean water to the atmosphere.[7]

The atmosphere provides an efficient transport route for micro-[5] and likely nanoplastic particles over long distances and to remote regions. Airborne nano- and microplastic particles may affect air quality, human health,[6] and atmospheric processes such as cloud formation,[7, 8] and the mechanisms and magnitude of these impacts remain unclear.

In this work we investigate the transfer of nanoplastic particles from water to air using a state-of-the-art sea spray simulation chamber[10] mimicking waters from fresh to slightly saline.

We characterize the particles in terms of size and chemical composition using scanning mobility particle sizing and pyrolysis gas chromatography coupled to mass spectrometry. Furthermore, we explore the hygroscopic properties of plastic particles containing salt using a hygroscopic tandem mobility particle sizer coupled to a high-resolution time-of-flight aerosol mass spectrometer. Our results will be discussed in the context of atmospheric processes, air pollution, global public health, and climate.

References

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[10] Christiansen, S. et al. Environmental Science & Technology **53**, 13107 (2019)

Human Aerosols and Drops and the associated transmission risk of respiratory pathogens

Gholamhossein Bagheri[1], Oliver Schlenczek[1], Laura Turco[1], Birte Thiede[1], Katja Stieger[1,2], Jana M. Kosub[3], Sigrid Clauberg[3], Mira L. Pöhlker[4,5], Christopher Pöhlker[5], Jan Molacek[1], Simone Scheithauer[3], Eberhard Bodenschatz[1,2,6]

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Understanding infection transmission between individuals, as well as evaluating the efficacy of protective measures, are key issues in pandemics driven by human respiratory particles. The key is a quantitative understanding of the size and concentration of particles exhaled and their variability across the size range for a representative population of all ages, genders, and different activities. Here we present data from 132 healthy volunteers aged 5 to 80 years, measured over the entire particle size range for each individual. Conventional particle spectrometry was combined with in-line holography under well-controlled conditions for common activities such as breathing, speaking, singing, and shouting. We find age to be the most important parameter for the concentration of small exhale particles (PM5), which doubles over a 7-year period in adolescents and over a 30-year period in adults. Gender, body mass index, smoking or exercise habits have no discernible effect. We provide evidence that PM5 particles originate from the lower respiratory from the larynx/pharynx, and >PM5 from the oral cavity. PM5 concentration can vary by one order of magnitude within a person, while inter-person variability can span two orders of magnitude, largely explained by difference in age. We found no discernible inter-person variability for particles larger than PM5. Our results show that cumulative volume of PM5 is 2-8 times higher in adults than in children. In contrast, number and volume concentration of larger particles, which are produced predominantly in the upper respiratory tract, is largely independent of age. Finally, we examined different types of airborne-transmissible respiratory diseases and provided insights into possible modes of infection transmission with and without several types/fits of face masks.

References

[1] Bagheri et al., Journal of Aerosol Science, accepted, 2022

Protons at the water surface: surface pH versus bulk pH

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Understanding the interfacial molecular structure of acidic aqueous solutions is important in the context atmospheric chemistry. We use surface-specific vibrational spectroscopy to probe the response of interfacial protons at the water–air interface. Combined with spectral calculations based on ab initio molecular dynamics simulations, the proton at the water–air interface is shown to be well-hydrated, despite the limited availability of hydration water. A substantial interfacial stabilization by −1.3 ± 0.2 kcal/mol is observed experimentally, in good agreement with our free energy calculations. The surface propensity of the proton can be attributed to the relatively poor hydrogen bonding between the hydrated proton and water molecules.

While these results reveal a very local enhancement of the proton concentration at the outermost few Ångströms of the water surface, a mesoscopic pH gradient from the surface to the core has previously been reported for aerosols. Here, we determine the pH distribution inside individual aerosol microdroplets using broadband coherent anti-Stokes Raman scattering micro-spectroscopy by quantifying the vibrational response of the conjugated acid-base pair of H_2 PO_4^- -- HPO_4^(2-). We find that the pH is essentially constant across the microdroplets with standard deviations below 0.2 pH units. The observation of a spatially homogeneous pH inside microdroplets, determined simply by the parent solution, resolves fundamental questions about the validity and sufficiency of methods applied for aerosol pH measurements.

Multi-Sensor Exposure Estimates to Help Better Understand Air Pollution Impacts

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Even with the knowledge that aerosols profoundy impact human health, progress towards reducing the impacts of air pollution is slow in many parts of the world. To mitigate the hazards of air pollution more effectively on health, we need to increase public understanding of the relationships between high pollution levels and resultant impacts. We usually inform the public of pollution levels by simply reporting the ambient concentrations and/or using air quality indicators that are associated with health qualifiers (good, unhealthy, etc.). However, there is a growing body of knowledge on how people at risk interpret, understand, and use information in making decisions. Information that is impact-based better supports improved decision making and planning. This is now well recognized in the weather services where there is a shift from “what the weather will be” to “what the weather will do.” There is also a need for improved estimates of exposure because public health organizations and professionals involved in these air pollution studies typically do not make use of the the many different observation streams. New sensor technology from space such as TropOMI, GEMS, MAIA, and TEMPO along with advances in ground-based measurements and global air pollution prediction and reanalysis products can provide high-resolution spatiotemporal information needed to produce better impact-based information. In this paper we present results of impact based analysis of projections of air pollution scenarious associated with different development pathways. We also proesent results for methods to produce high resilotion estimates of pollution sitributions for use in health studies.

Peroxy radicals really are at the center of the universe

Neil M. Donahue[1] and CLOUD Science Team[2]

[1]Carnegie Mellon University, Pittsburgh, USA
[2] CERN and CLOUD institutes, Geneva, Europe and USA

Peroxy radicals, including hydroperoxyl radical (HO_2) and organoperoxy radicals (RO_2) have long been recognized as central players in tropospheric chemistry. In general, RO_2 are produced immediate after the onset of photochemical oxidation of any organic compound in the atmosphere – any carbon centered radical will immediately react with molecular oxygen to form RO_2. In the atmosphere, RO_2 have lifetimes of 1 – 1000 s and their branching defines the chemical environment. The major focus has been on whether RO_2 react with HO_2 or NO, which loosely defines "low NO_x" and "high NO_x" conditions. Conveniently, the rate coefficients for those reactions are similar and do not depend strongly on the organic group, R. Roughly 1 in 30 collisions with either radical result in a reaction, independent of temperature (for the most part). The HO_2:NO ratio is thus critical. However, there is a growing recognition of 3 surprises: some RO_2 can undergo rapid isomerization (internal H-atom abstraction) followed by O_2 addition to reform oxygenated RO_2 (this is autoxidation); some RO_2 + RO_2 reactions can be very fast, especially when the R group is highly oxygenated (electron withdrawing functional groups near the OO• may be key); some RO_2 + RO_2 reactions produce organic peroxides (ROOR + O_2) with high yields, also apparently when the R group is highly oxygenated. These three novel factors combine to reveal a fascinating chemical landscape coupling different organic compounds that may have changed dramatically through industrial times, may largely define chemical conditions in gigacity environments, and whose evolution along future decarbonization trajectories may be very important to air quality and climate consequences of those policies.

The Twin Forcing Agents of Urban Climates: Impacts and Perspectives on Future Directions

M. Georgescu[1]

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In this talk, I will examine the urban climate system by highlighting the separate and distinct contributions to urban climate arising from greenhouse gas induced climate change and the built environment. I will review recent physically-based regional climate modeling work examining projected effects on near-surface temperature and precipitation, including means and extremes, associated with future urbanization, large-scale climate change and their interaction across the continental US. I will discuss how detrimental impacts on temperature from these aforementioned forcing agents could be reduced through application of a suite of biophysical approaches. I will discuss future implications for extreme heat across US cities that includes the twin forcing agents of urban infrastructure and climate change, and examine the potential to reduce heat exposure from adaptation and mitigation. I will conclude with a view of my perspectives on the future of urban climatology research and its contextual relationship with the recent proliferation in data sources, exceptional growth in computational capabilities, and systems-thinking approaches that aim to fundamentally transform our approach to understanding of urban climate dynamics.

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Natural Aerosol Impacts on Air Quality and Climate

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Aerosol particles are emitted and produced from myriad sources, both anthropogenic and natural. Natural aerosol sources provide a key baseline for both aerosol climate forcing and fine particulate matter ($PM_{2.5}$) exposure. I will discuss how these sources of PM are increasingly important to understand and quantify in light of strengthening WHO guidelines on $PM_{2.5}$ exposure [1]. I will briefly highlight three relevant examples from our recent work. First, how the fate of biogenic peroxy radicals (RO_2), which are the precursors to biogenic secondary organic aerosol (SOA) varies globally and has changed from pre-industrial to present-day. Second, how attributing smoke emissions and exposure by ignition type in the United States, suggests that little fire exposure in this region is wholly "natural" [2]. And finally, how expanding our representation of DMS chemistry in a global earth system model alters the formation of sulfate and its radiative impacts from pre-industrial to present day [3].

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Chemistry in nanometer particles: Laboratory studies on particle size-dependent aerosol chemistry

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Currently, the formation of extremely low-volatile organic compounds required for nanometer particle growth is mainly discussed based on the formation of HOMs by gas-phase chemistry (autoxidation). However, the nanometer particles themselves provide a unique nanoscale chemical environment that can influence the chemical reactions within the newly formed condensed phase. Beyond pure gas-phase reactions, several formation pathways of very low-volatility organic compounds exist. One is heterogeneous oxidation reactions, e.g. at the particle surface, another is the formation of organic salts and finally oligomerization reactions in the condensed phase. Of particular interest are reactions that preferentially occur in the smallest particles, since it is these particles that require matter for their growth into size ranges where they are not rapidly lost by coagulation. In these cases, particle size-dependent chemical reactions may play a critical role in the life cycle of atmospheric aerosols by bridging the gap between the initial formation of particle embryos and their growth into size ranges where their survival probability is greater and on which cloud droplets can eventually form. Here we report the results of laboratory experiments on particle size-dependent reactions, e.g., model reactions on the influence of pressure on oligomerization reactions (Diels-Alder reactions), heterogeneous oxidation processes, and condensation reactions in which volatile products are formed. All of these reactions could or should be influenced by the curvature of the surface of organic aerosol particles and accordingly can show a size dependency. The studies were carried out in laboratory-scale flow reactors at the University of Mainz. In particular, mass spectrometric analytical techniques such as UHPLC-ESI-Orbitrap MS and GC-Orbitrap MS measurements as well as on-line CI-Orbitrap-MS studies were used. Size-dependent chemical reactions are shown to play a role in the size range between 30 and 100 nm, and thus could be important for the growth of freshly formed particles up to a size where they can act as cloud condensation nuclei (CCNs).

Particle filtration efficiency and breathability of commonly used facial masks

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Airborne transmission of SARS-COV-2 virus has been recognized as an important pathway for infection. In many countries wearing a mask has become one of the prevention measures. Good ventilation is rather a recommendation and not a requirement. Social distancing and disinfecting hands are important, but not effective against the airborne transmission.

Even though, the masks are compulsory in public places the quality of masks are poorly understood by public, and the packaging of the masks is often not informative. To improve the general knowledge on particle filtration efficiency (PFE) and breathability of the masks, we called up a citizen campaign to study commonly available and used masks in Estonia.

We received 139 different masks, and all were analysed for PFE for sizes of 10nm to 10mm and for the pressure drop, the breathability. Some selected masks were also treated with hot water and washed with a washing machine to study the performance changes with different heat-based disinfection treatments.

Results reviled substantial differences between the masks. The results were published in freely accessible website (https://atmos.ut.ee/en/masks)

In the presentation the results are examined in more details and benefits of mask wearing are discussed.

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Current understanding on atmospheric new particle formation

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We summarize our latest findings on atmospheric new particle formation (NPF) based on atmospheric observations and model simulations. Our first investigation was a closure study on sub-6 nm atmospheric aerosol particles and clusters, which showed that present observations are able to detect a major fraction of existing atmospheric clusters [1]. Our second finding, based on long-term measurements in four different environments, was that atmospheric NPF tend to occur also on days characterized traditionally as non-event days (days with no observed NPF). We termed this phenomenon “quiet NPF” and showed that it may have a significant contribution to the production of secondary particles in the atmosphere [2]. Third, we investigated the growth of newly-formed particles into sizes relevant for climate and air quality using box model simulations that were constrained with atmospheric observations in two very different environments: 1) Beijing, a polluted megacity in China and 2) SMEAR II station, a boreal forest site in Finland. Our simulations for Beijing showed that NPF in capable of giving large contributions of haze particle mass and number concentrations [3]. The results indicate that reducing primary particle emissions may not decrease PM pollution effectively in heavily polluted environments until simultaneous emission reductions will be made for precursor gases responsible for NPF and subsequent particle growth. At SMEAR II, we used simulations to investigate the role of NPF in the Continental Biosphere-Atmosphere-Cloud-Climate (COBACC) feedback mechanism [4]. We found that outside the late autumn and winter periods when NPF tends to be rare at SMEAR II, NPF gives a dominant contribution to both condensation sink and cloud condensation nuclei concentration – the two most relevant quantities in the COBACC feedback mechanism. We estimated that the same conclusion is likely to hold over large regions in the boreal forest zone. As a side product of our observations, we found a surprisingly low variability in growth rates of newly formed particles in both Beijing and SMEAR II. This indicates a potentially important role of multi-phase reactions causing the bulk growth of newly formed atmospheric particles – a phenomenon that needs to be investigated in more detail in future.

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Perspective on atmospheric particle formation from the CERN CLOUD experiment

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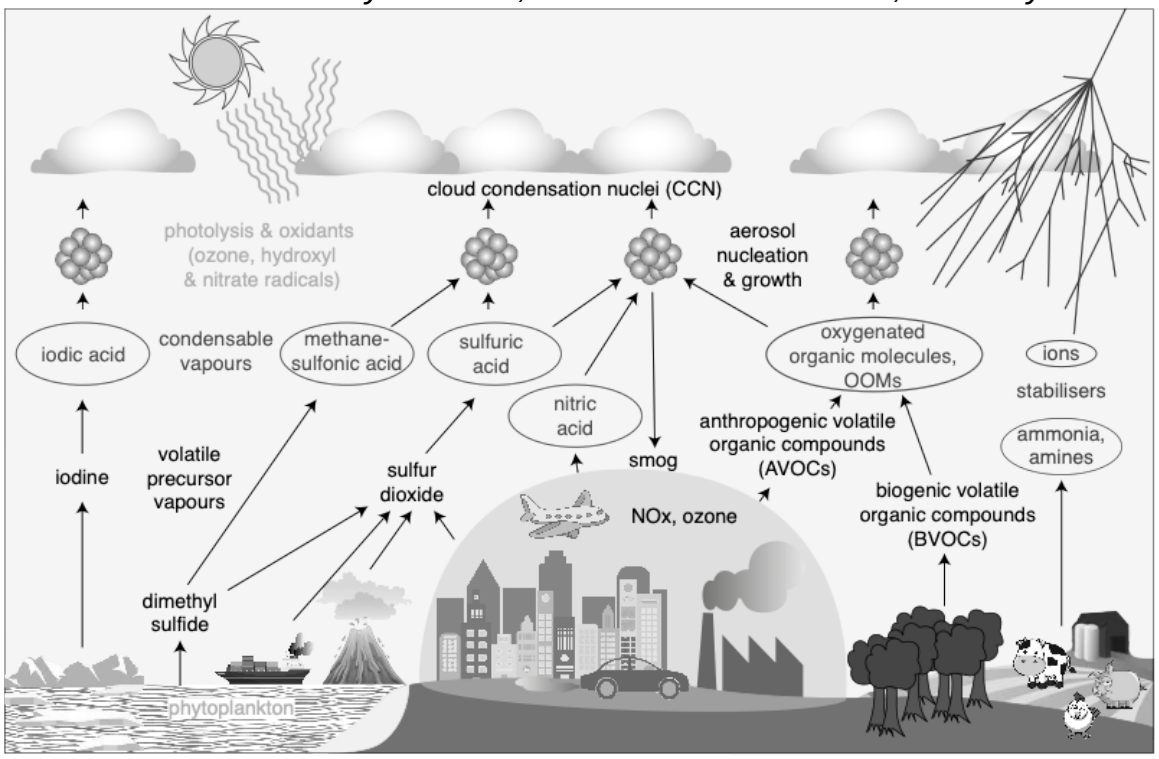

Fig. 1 Current understanding of the main vapours and processes responsible for new particle formation and growth in the atmosphere. *Condensable vapours comprise sulfuric acid, methanesulfonic acid, oxygenated organic molecules (OOMs), iodic acid and, under certain conditions, nitric acid. Stabilisers, which reduce evaporation of embryonic molecular clusters, comprise ammonia, amines and ions from galactic cosmic rays. Atmospheric oxidants - especially hydroxyl radicals and ozone - transform volatile precursor vapours such as sulfur dioxide, dimethyl sulfide, organic compounds or iodine into ultra low volatility vapours.*

The understanding of atmospheric new particle formation (NPF) has been transformed over the last few years. Most of the main chemical species and mechanisms responsible for atmospheric NPF and growth may have now been identified (Fig.1), but their rates under different atmospheric conditions and the interplay between these vapours is only partially characterised so far. This paper will present a perspective on atmospheric new particle formation and growth from experiments at the CERN CLOUD chamber[1] over the last thirteen years.

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Interactions of boundary layer dynamics, urban climate, and air quality in the Chinese Gigacity

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The urbanization and industrialization of China in the recent decades have been unprecedented in the human history. The clusters of megacities in northeast China could be classified as one huge urban area – a Gigacity[1], defined roughly by the lines Beijing – Xian – Shanghai – Beijing. Air pollution is one of the most pressing environmental problems in terms of human health in China. Therefore, it is crucial to understand the factors and the complex interactions affecting the near surface air quality.

Even though the effect of haze on the reduction of urban boundary layer height (BLH) is rather well-known phenomena, there are still significant knowledge gaps in the seasonal variation of the effect of haze on the BLH and its driving forces. We analyzed the BLH utilizing observations from the Beijing University of Chemical Technology (BUCT) for the year 2018, supplemented with simulations using Surface Urban Energy and Water Balance Scheme (SUEWS[2]). The effect of haze on the BLH was the highest in spring and autumn, while having substantially smaller effect in summer. Based on our results, this is presumably related to the seasonal variation of the magnitude of the surface heat fluxes.

We examined the effect of the few most regularly used urban morphological characteristics on spatial variation of air quality in downtown Nanjing. We performed this study in real urban environment using continuous observations from 1 December 2019 to 29 February 2020 in 31 study sites with highly varying urban characteristics covering the whole range of urban densities typically found in cities. The effect of nearby trees was identified to be the most important urban morphological characteristics defining the near-surface pollutant concentrations in street canyons and the height normalized roughness length as the second most important.

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Global impacts of particulate air pollution on public health

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The long-term exposure to fine particulate matter (PM2.5) poses a main public health risk. Our group has developed a data-informed global atmospheric model to compute the exposure to particle concentrations, being attributed to different source categories. We apply exposure-response functions to assess the health burden, including excess mortality. It appears that black carbon and anthropogenic organic aerosols are more harmful than other PM2.5 constituents, related to their high oxidative potential upon inhalation. Considering that these species may be about twice as hazardous as other PM2.5 compounds, domestic energy use emerges as a leading cause of mortality attributable to PM2.5, notably in Asia and Africa, while in Europe and America the use of fossil fuels predominates. Recently, we computed the global concentrations of ultrafine particles (UFP, <0.1 μm) at high spatial resolution (10km). UFP are of special interest, as they can translocate into the blood stream and trigger systemic inflammatory responses, important for cardiovascular disease. From the global exposure to UFP we estimated the cardiovascular disease incidence. Preliminary results indicate that a large fraction of cardiovascular disease, previously attributed to PM2.5, may actually result from UFP. Globally, the mean loss of life expectancy from air pollution surpasses that of many known health risk factors such as infectious diseases, and is comparable to that of tobacco smoking.

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Air quality, climate changes and their intertwined relationships in Asia

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Air pollution and climate changes have emerged as major concerns to the well-being of mankind, which are particularly acute in Asia centered in China associated partially with its dense population, fast development, and special topography. Moreover, the two are intertwined that could amplify their detrimental effects. Anthropogenic aerosols, particulate pollutants in the atmosphere, can affect the atmospheric processes in numerous ways. While mounting evidence have been presented to illustrate their impact on radiation, temperature, precipitation, severe storms, it has been a daunting task to identify, understand and quantify the various effects, working mechanisms and connections. Few places are more ideal than China to unravel the complex relationships where pollution has been severe and changed rapidly with strong long-term up- and down-trends. By means of field experiments, satellite remote sensing and modeling, our team has tackled with the complex problems in a systematic approach. I will present some major findings of these studies.

An invited talk at the WE-Heraeus Seminar on " Aerosols, Health and Climate: Gigacity and Future", 20 to 24 March 2023 at the Physikzentrum Bad Honnef of the German Physical Society

Aerosol impact on climate by impacting warm and mixed-phase clouds

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Clouds are not only fascinating to observe for their myriad of shapes, but are also scientifically challenging because their formation requires both knowledge about the large-scale meteorological environment as well as knowledge about the details of cloud droplet and ice crystal formation on the micro-scale. The ice phase in clouds remains enigmatic because ice crystal number concentrations can exceed the number concentrations of those aerosol particles acting as seeds for ice crystals (so-called ice nucleating particles, INPs) by orders of magnitude. Aerosol particles can scatter and absorb solar radiation and with that cause a cooling, that partly offsets the greenhouse gas warming. Aerosol particles also influence the microphysics of clouds by acting as cloud condensation nuclei (CCN) and INPs. An increase in aerosol particles will lead to more and smaller cloud droplets for a given cloud water content, which leads to an increase in cloud albedo (Twomey effect).

The magnitude and geographical distribution of the cooling caused by aerosol particles themselves and by aerosol-cloud interactions are much more uncertain than the greenhouse gas warming because aerosol particles have localized sources and sinks and only have an atmospheric residence time of days to weeks.

The 2-months effusive volcanic Holuhraun eruption in 2014 in Iceland has been used to understand and quantify the influence of aerosol perturbations on clouds using satellite data combined with machine-learning. Chen et al. [1] concluded that the increase in cloud coverage during this time is more important for the effective aerosol radiative forcing than the increase in albedo from the Twomey effect.

Aerosol effects on mixed-phase clouds (clouds containing cloud droplets and ice crystals at the same time) are more complex than in warm clouds because aerosol particles can act both as CCN and as INPs and more microphysical pathways exist. One suggestion, how mixed-phase clouds could be modified in order to alleviate some of the global warming has recently been suggested [2].

In this lecture, the progress that has been made in understanding the influence of aerosol particles on clouds and climate will be addressed.

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Effects of air pollution on cardiovascular disease

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Ambient air pollution is a leading cause of non-communicable disease globally. The largest proportion of deaths and morbidity due to air pollution is now known to be due to cardiovascular disorders. Several particulate and gaseous air pollutants can trigger acute events (e.g. myocardial infarction, stroke, heart failure). While the mechanisms by which air pollutants cause cardiovascular events is undergoing continual refinement, the preponderant evidence support rapid effects of a diversity of pollutants including all particulate pollutants (e.g. course, fine, ultrafine particles) and gaseous pollutants such as ozone, on vascular function. Indeed, alterations in endothelial function, a subclinical marker for atherosclerosis, seem to be critically important in transducing signals and eventually promoting cardiovascular disorders such as hypertension, diabetes, and atherosclerosis including arterial hypertension, myocardial infarction, heart failure, stroke and arrhythmia [1]. Here, I will provide an updated overview of the impact of particulate and gaseous pollutants on endothelial and cardiovascular function from human and animal studies. The evidence for causal mechanistic pathways from both animal and human studies that support various hypothesized general pathways and their individual and collective impact on vascular function is highlighted. I will also discuss effective personal mitigation maneuvers to effectively fight air pollution induced cardiovascular disease [2]. To this end I will conclude by an exhortation for formal inclusion of air pollution as a major risk factor in societal guidelines and provision of formal recommendations to prevent adverse cardiovascular side effects attributable to air pollution.

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Strong control of aerosol-cloud interactions by emissions from the boreal forests

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Boreal forests are a carbon sink and contributes to the formation and growth of secondary organic aerosols through oxidation products from their biogenic volatile organic compound emissions. The overall influence of these secondary aerosol particles to the global climate system are poorly quantified. We explored how condensable organic aerosol precursors were transported and transformed during their lifetime in the boreal atmosphere and we were able to connect the aerosol processes and aerosol-cloud interactions in warm clouds [1]. The interaction is highly dynamical. We combined 8 months of in-situ and ground-based remote sensing data from SMEAR II station in Hyytiälä, Finland obtained during BAECC campaign [2] in 2014. We observed a substantial increase in aerosol number and mass concentrations over the forest corresponding to one to three days after the marine air enters the boreal forest atmosphere. The findings are consistent with secondary organic aerosol formation and, together with evapotranspiration, are associated with changes in the radiative properties of warm, low-level clouds [1]. Such feedbacks between boreal forest emissions and aerosol–cloud interactions over timescales of several days suggest that the boreal forests have the potential to influence climate change on a continental scale.

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Research into Aerosols, Films and wider Atmospheric Chemistry.

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Organic aerosols in indoor and outdoor air often contain both hydrophilic and hydrophobic components, but the nature of how these compounds are arranged within an aerosol droplet remains largely unknown. We have developed a range of novel experimental approaches to study molecular processes at air–water and air–solid interfaces as well as in levitated aerosol particles and droplets e.g. demonstrating that acoustic trapping[1] can be used to levitate and manipulate droplets of soft matter, such as lyotropic mesophases formed from self-assembly of different surfactants and lipids, which can be analysed in a contact-less manner by X-ray scattering in a controlled gas-phase environment. We have shown[2,3,4,5] that fatty acids in proxies for atmospheric aerosols from cooking and sea spray self-assemble into highly ordered three-dimensional nanostructures that may have implications for environmentally important processes in indoor and outdoor air. We investigated the atmospheric lifetimes of major components of cooking emissions with unique surfactant properties that may enable them to survive in the urban atmosphere for an extended period in self-assembled molecular matrices.[2] We have followed the kinetics of sub-µm self-assembled films using Small-Angle X-ray Scattering (SAXS) validated by simultaneous Raman microscopy.[3] Our experiments demonstrated that self-assembled material is retained even upon extended exposure to high ozone concentrations. In the indoor space, we have investigated self-assembly of films on solid substrates („window grime“).[4] Kinetic modelling[5,6] linked to studies of multi-component cooking emission proxies was used to estimate the potential impact on urban air quality of these extended lifetimes of reactive species that have previously been assumed to be short-lived and removed rapidly from the atmosphere. We suggest that lyotropic-phase formation likely occurs in the atmosphere, with potential implications for residence times and other aerosol characteristics both in indoor and outdoor air.

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Physical, chemical, and biological multiphase processes influencing climate and health in the Anthropocene

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Multiphase processes involving gases, liquids, and solid or semi-solid matter play important roles in the Earth system as well as in living organisms. Some of these processes are particularly important for climate change and public health in the Anthropocene, i.e., in the current era of globally pervasive and rapidly increasing influence of human activity on planet Earth. For example, the interplay of water vapor with aerosol particles influences the formation of clouds and precipitation, which are essential for the Earth's energy balance and hydrological cycle. We quantify aerosol properties and their interactions with clouds, precipitation, and radiation in contrasting clean and polluted environments ranging from near-pristine environments in remote marine regions and the Amazon rainforest (blue vs. green ocean) to densely populated Asian megacity regions. In particular, we focus on the interactions of carbonaceous particulate matter including black carbon (soot), secondary organic aerosols, and primary biological particles (bioaerosols). We investigate how phase changes and related parameters influence the atmospheric transport and transformation of hazardous pollutants like polycyclic aromatic compounds as well as natural contaminants like allergenic proteins and their nitrated, oxygenated, and oligomerized derivatives. Moreover, we study how aqueous-phase chemistry affects the atmosphere–biosphere exchange of reactive nitrogen. With regard to aerosol health effects, we study how gaseous pollutants and components of fine particulate matter (PM2.5) generate reactive oxygen and nitrogen species causing oxidative stress, inflammation, and chemical modifications of proteins and other biomolecules, which may lead to non-communicable respiratory, cardiovascular, and allergic diseases. In the coronavirus pandemic, we also assessed how the transmission of infectious diseases such as COVID-19 by respiratory aerosols and droplets can be effectively reduced by protective measures like masking, distancing, and ventilation.

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Recent findings on air pollution sources, processes and links to health in Indian and Chinese cities

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Northern India and Eastern China are among the most polluted areas in the world. The population in mulitple megacities in these regions suffer from very high exposure to air pollution. Novel techniques allow for better source apportionment of the air pollution sources including particulate matter (PM) and volatile organic compounds. The importance of PM inorganic secondary species, the organics, black carbon and trace elements have been studied in great detail. Regarding the organics it is known since a while that the chemically processed oxygenated organic aerosols are a very important fraction of the organic aerosols even in winter in megacities [1[. Primary organics include typically contributions from solid fuel combustion, traffic, cooking. The formation processes of oxygenated organic aerosols are not yet fully understood. Classic photochemical formation of secondary organic aerosols from volatile organic compounds are surely important but other processes also play a role including night-time chemistry, aqueous phase reactions, light induced reactions which will be discussed. Recently it was shown that organics, especially secondary organics are harmful to the population due to reactive oxygen species (ROS) [2]. Besides organics also some metals are crucial for the adverse ROS concentrations. Therefore it is important to also understand the trace metal composition and sources. The links of ROS to chemical composition in these urban areas will be discussed. We propose that a combination of real-time instruments including real-time source apportionment is implemented in such cities in combination with ROS measurements to be able to implement measures in timely manner and to study in more detail the health impacts.

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Insights into aerosol-cloud-climate interactions in environments with limited human influence

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Aerosol-cloud interactions are currently among the key uncertainties hampering accurate estimates of the contribution of anthropogenic aerosols on radiative forcing during the industrial time. Understanding aerosol loadings originating from various natural sources, on the other hand, is necessary to estimate the preindustrial baseline conditions that provide the reference point for our forcing estimates. In my presentation, I will discuss some of our recent work investigating the key processes controlling aerosol size distribution, composition and hence Cloud Condensation Nuclei (CCN) concentration in environments with limited human influence (boreal and tropical forests, and the Arctic). The studies covered utilize methodologies ranging from in-situ and remote sensing observations (both long-term and campaign-wise) to process-based, cloud-resolving and Earth System Modeling. The results highlight the need for accurate predictions of aerosol size distribution evolution and consistent, yet simple enough, treatment of all the relevant steps from emission of precursor vapors to aerosol dynamics, aerosol-cloud -interaction and precipitation formation and effects on aerosol loadings.

Vertical profiling of aerosol particles in Arctic regions

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Aerosol particles are involved in numerous feedback processes that contribute to the warming of the Arctic, which is occurring twice to three times as fast as the global average. Due to the lack of continuous and vertical measurements, the knowledge about the distribution, transport, sources and sinks of aerosol particles in the Arctic is still poor [1]. Vertical measurements are mainly limited to ground-based locations, only a few campaigns did vertical measurements in the central Arctic, based on ice floes or research vessels.

This study presents recent results from vertical measurements of aerosol particles in the Arctic boundary layer. We present UAS measurements from Ny-Ålesund (2018) and tethered balloon measurements taken on an ice floe during MOSAiC [2][3] (2020) as well as in Ny-Ålesund (2021/22).

The UAS ALADINA is equipped with two particle counters for total number concentration from 3 and 12 nm and one optical particle counter for particle size distributions above 300 nm [5]. The tethered balloon system BELUGA is flexible to carry various different measurement platforms at the same time or subsequently. The cubic aerosol measurement platform CAMP was developed for operation at BELUGA to measure aerosol parameters, equipped with instruments similar to ALADINA and an additional absorption photometer [4].

In total 29 flights were performed with ALADINA in April/May 2018 with a total duration of 50 flight hours up to heights of 850 m below cloud ceiling or at cloud-free cases. The profiles show a clear dependence on the surrounding orography but also air mass history. A few cases of local new particle formation were found.

During MOSAIC leg 4 in July 2020, 36 aerosol profiles were measured using the BELUGA-CAMP system most of them under cloudy conditions. Here, a detailed analysis in connection with boundary layer parameters will be shown.

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Atmospheric Aerosol Chemistry: From the Boreal Forest to Gigacities

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Despite much effort in the past decades, uncertainties in both climate impacts and health effects of atmospheric aerosols remain large. During the last 25 years, aerosol mass spectrometry (AMS) has shown that sub-micron aerosol chemical composition is roughly 50:50 inorganic and organic worldwide, with secondary highly oxidized organics dominating the latter. Parallel application of chemical ionization mass spectrometry (CIMS) has detected molecular cluster ions involved in atmospheric nucleation, including detection of highly oxidized multifunctional (HOM) organics in the gas phase; which also are a significant contribution to secondary organic aerosol in both clean and polluted environments. These results will be discussed in the context of their impact on air quality and climate; from the boreal forest to Chinese megacities.

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Formation and Human Health Impacts of Ultrafine particles

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Airborne particulate matter (PM) or aerosol (particles suspended in air) exerts profound impacts on the atmosphere-earth system, ranging from air quality, ecosystems, weather, to climate. A large fraction of the tropospheric aerosol populations is produced from new particle formation (NPF) under various environments. Notably, the atmospheric effects of ultrafine particles (UFPs) are distinct from those of larger particles (e.g., fine and coarse particles). Elevated UFP levels have been attributed to substantially enhanced convection and precipitation. Because of their ability to enter extra-pulmonary tissues, UFPs have been linked to strong adverse human health effects. Beside the lung, many other organs are also subjected to the impacts of UFPs. The nanotoxicity of UFPs overproduces reactive oxygen species (ROS) with permanent oxidative damage to lipids of cellular membranes, proteins, and DNA and cell failure for normal physiological redox-regulated functions, leading to metabolic, immune, and cardiovascular dysfunctions. Exposure of UFPs to vulnerable individuals is especially pronounced, such as for pregnant women. Currently, there exists a high degree of exposure misclassification for UFPs, which are unregulated in the available air quality standards. In this talk, I will present results relevant to investigations of formation and health impacts of UFPs. I will present research that discovers a unique onset for the occurrence of NPF under urban conditions. In addition, I will discuss work employing animal models to demonstrate that prenatal exposure to UFPs leads to stillbirth, low birth weight, and organogenesis as well as respiratory, and cardiometabolic dysfunctions relevant to both conceptus and postnatal growth and development.

Abstracts of Posters

(in alphabetical order)

Exploring low-level haze formation mechanisms using customized drone setup at Delhi

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During post-monsoon and winter seasons, Delhi experiences severe hazy events impacting climate, health and economy. Recently, a few ground-based studies were conducted in Delhi during hazy periods to enhance our understanding on haze formation mechanisms [1]. However, due to lack of airborne observations in lower boundary layer (~100 m above ground level), there is a significant knowledge gap regarding vertical information of air pollutants during hazy period and, thus, low-level haze formation at Delhi. Therefore, an airborne campaign using customized drone set-up was conducted at Indian Institute of Technology (IITD), Delhi to explore the possible mechanisms of early morning haze formation (moderate haze, during pre-monsoon season) from March 12-23, 2021. The drone set-up carried a payload for measuring vertical distribution of air pollutants such as PM_1, $PM_{2.5}$ and equivalent Black Carbon (eBC) mass concentrations and meteorological parameters. The ground-based instruments (aerosol chemical speciation monitor (ACSM), aethalometer (AE-33) etc.) at IITD site provided ground-based chemical composition, eBC mass concentration to complement the vertical measurements. The PM_1 mass concentrations were found to be high while eBC mass concentrations were low at 100 m a.g.l. respectively when compared to ground level during hazy mornings. The PM_1 aerosol chemical composition revealed organic concentration (~40 µg/m^3) and high chloride concentration (~30 µg/m^3) respectively at ground on alternate mornings (hazy). In addition, the vertically measured data was compared to WRF-Chem model. The results showed large underestimation in PM_1 mass concentration during hazy mornings simulated from WRF-Chem when compared to drone-based observations. This could be due to assumption of uniform data throughout the vertical column in the absence of vertical air pollutants information. The campaign findings suggest that further detailed vertically resolved (radicals, volatile organic compounds (VOCs), aerosol composition, mixing-state, size etc.)) data using drone set-up is required to predict extremely severe haze during post-monsoon and winter seasons.

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Aerosols and the plight of the exposed: understanding personal exposure through microscale air pollution research

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Air pollution is personal. The human's exposure to harmful air pollutants is a result of the combination of the unique behavior of the individual and his/her interaction with the environment. Hence, understanding personal exposure requires a microscale approach. In this seminar, I will be presenting our microscale urban air pollution research performed using mobile platforms and the 5 main applications that we have explored. This spans from development of measurement strategies for better representation of personal exposure [1] to exploratory pursuits of transdisciplinary studies in megacities [2]. In the past decade, we have performed mobile measurements of particulate aerosols with a focus on how they vary in space, effectively mapping air pollution and identifying hotspots in key areas of Manila [3], Leipzig [4], Rome [5], and La Paz. Since the instruments aboard the mobile platform are scientific-grade, we also employ it to validate a low-cost PM network currently monitoring residential wood burning Leipzig. Further developments have led to a platform that provides an estimate of the black carbon particles deposited in the respiratory tract. The deposition dose has been determined for large cities: in a high-altitude region of La Paz [6] and at the sea level megacity of Manila [7]. Because of its versatility, the mobile platform has also been used for indoor air quality research: exposure of Rwandan women to wood smoke during cooking and penetration of mineral dust in Cape Verdan households during the dust season. Finally, its simplicity enabled us to use it as the main platform for engaging the public in knowledge transfer projects in Germany. With this mobile platform, in combination with our expertise on more elaborate aerosol measurements strategies, the potential is ripe for interdisciplinary and transdisciplinary studies focused on increasing our understanding on the health effects of air pollution and improvement of air quality in cities.

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Machine Learning-Based Prediction of the Glass Transition Temperature

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Knowledge of the glass transition temperature of molecular compounds in atmospheric aerosol particles is important for estimating their viscosity [1], which directly influences chemical reaction kinetics and phase state [2]. While there is a great diversity of organic compounds present in aerosol particles, experimental glass transition temperatures are known of only a minor fraction of them [3]. Therefore, we have developed a machine learning model in Python designed to predict the glass transition temperature of organic molecular compounds based on molecule-derived input variables. The extremely randomized trees (extra trees) procedure was chosen for this objective. Two approaches using different sets of input variables were followed. The first one uses the number of predefined functional groups present in the compound, while the second one generates descriptors from a SMILES (Simplified Molecular Input Line Entry System) string. For improved results both approaches can be combined with the melting temperature of the compound as an additional input variable, if known. The results show that the SMILES-based predictions had a slightly lower mean absolute error (MAE), but both approaches had a similar MAE of about 11-13 K. Furthermore, we also show that its performance exceeds that of previous parametrizations developed of this purpose and also performs better than existing machine learning models. We believe that this model is a powerful tool for many applications in atmospheric aerosol science and material science.

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A global climatology of ice-nucleating particles under cirrus conditions derived from model simulations with MADE3 in EMAC

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Atmospheric aerosols can act as ice-nucleating particles (INPs) and thereby exert important influences on the formation and the microphysical properties of cirrus clouds and in turn induce distinct climate effects [1, 2]. In laboratory experiments several types of aerosol particles have been identified as efficient INPs under cirrus conditions [3]. However, the knowledge about the global atmospheric distribution of INPs is still limited and consequently the understanding of their climate impacts is highly uncertain. We perform model simulations with a global aerosol-climate model coupled to a two-moment cloud microphysical scheme and a parametrization for aerosol-induced ice formation in cirrus clouds and present a global climatology of INPs in the cirrus regime [4]. This novel INP climatology comprises, in addition to the broadly considered mineral dust and soot INPs, also crystalline ammonium sulfate and glassy organic particles. By coupling the different INP types to the microphysical cirrus cloud scheme, their ice nucleation potential under cirrus conditions is analyzed, considering possible competition mechanisms between different INPs. The simulated INP number concentrations range from about 1 to 100 L^{-1} and agree well with in-situ observations and other global model studies. Our model results show that glassy organic INP concentrations are mostly low in the cirrus regime, suggesting a small climate impact. On the other hand, crystalline ammonium sulfate often shows large INP concentrations, resulting in a strong potential to influence ice nucleation in cirrus clouds, and should be taken into account, in addition to the effects of mineral dust and soot, in future model applications.

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KM-SUB-LUNG – A multiphase kinetic model untangling the chemical and biological contributions of air pollution to oxidative stress in the lungs

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Air pollution is a major risk factor for public health and epidemiological studies identify fine particulate matter (PM2.5), ozone (O3), and nitrogen oxides (NOx) as the most hazardous pollutants.[1,2] Oxidative stress in the lung due to an excess presence of reactive oxygen species (ROS) is the leading hypothesis for the mechanism behind the adverse health effects of air pollution.[3] Accordingly, measurements of the production of ROS by air pollutants are an increasingly popular means to assess their toxicity, for example in assays determining the so-called oxidative potential (OP) of PM2.5.[4] ROS, however, are a heterogeneous group of several radical and non-radical species, which are already present and released in significant quantities in the human body.[5] Furthermore, numerous defense mechanisms for oxidative stress protect tissues through enzymatic and antioxidant reactions, which are often not considered in OP assays. Here, we use a multiphase chemical kinetics model of the human respiratory tract, KM-SUB-LUNG, to quantify and compare the effects of gas-phase and particulate pollutants on the production of ROS and oxidative damage in the epithelial lining fluid of the lung.[6] The model aims at bridging the gap between laboratory investigations of air pollutant properties and the epidemiological evidence for air pollution health effects by providing physiological, quantitative metrics for air pollution toxicity. Our findings suggest that the adverse health effects may not be primarily related to direct chemical production of ROS and the OP of air pollutants, but rather to the conversion of peroxides into more reactive species such as the hydroxyl radical, or the stimulation of biological ROS production. The analysis highlights remaining uncertainties in the relevant physical, chemical and biological parameters, suggesting a critical reassessment of current paradigms in elucidating and mitigating the health effects of different types of air pollutants.

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New-Particle Formation in polluted environment: A comparison between Beijing and the Po Valley.

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Intense and frequent new particle formation (NPF) events have been observed in polluted urban environments, yet the dominant mechanisms are still under debate. To understand the key species and governing processes of NPF in polluted urban environments, we conducted comprehensive measurements in downtown Beijing and we compare them with measurement from the Po Valley in Northern Italy.

We performed detailed analyses on sulfuric acid cluster composition, as well as the chemical and physical properties of oxidized organic molecules (OOMs). Our results demonstrate that the fast clustering of sulfuric acid (H2SO4) and base molecules triggered the NPF events. Even though the mechanism is relatively similar it seems that the amine concentration in the Po valley is a bit lower and this affect the NPF mechanism.

Additionally, for the Beijing case, the synergistic role of H2SO4, base species, and OOMs in NPF is likely representative of polluted urban environments where abundant H2SO4 and base species usually co-exist, and OOMs are with moderately low volatility when produced under high NOx concentrations[1].

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Cool cloud chemistry for climate: Organic aerosol's ability to nucleate ice in mixed-phase clouds

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Aerosol-cloud interactions play a key role in the earth's energy budget yet contribute to a large uncertainty in radiative forcing in climate models. My group is interested in organic aerosols and its chemical composition to inform on the freezing of supercooled water droplets in mixed-phased clouds. To explore the mechanism of freezing of organic matter in cloud water conditions, we've developed drop freezing techniques relevant for immersion freezing.(*1*) We've also been exploring different types of samples: field-collected dissolved organic matter from lakes and rivers relevant for lake spray aerosols,(*2*) humic substance standards,(*2*) lignin as a subcomponent of biomass burning organic aerosol,(*3*) firewood smoke, ammonium sulfate methyl glyoxal brown carbon solutions, lab-generated secondary organic aerosols,(*4*) soil dust and proteinaceous material. These different samples are demonstrating that the size of the macromolecules and their functional groups are playing competitive roles in the ability of organic matter to template ice. In all, organic aerosols can alter the aerosol-cloud radiative effects by modifying the supercooled liquid water-to-ice crystal ratio in mixed-phase clouds with implications for cloud lifetime, precipitation patterns and the hydrological cycle.

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Long-term reduction of fine particulate matters in Beijing after the COVID lockdown: an investigation based on more than 3-year observations

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Abstract: Air pollution characterized by high fine particulate matter ($PM_{2.5}$) loadings, as one of the main environmental challenges, has attracted persistent attention in Chinese megacities. The COVID lockdown in Beijing in 2020 was found to result in a rapid and substantial reduction in pollutant emissions. In contrast, enduring effects on air quality and human health, potentially due to the significant alterations in human activities during the COVID pandemic, have not yet been investigated. In this study, based on long-term FLEXPART simulations and more than 3-year field measurements (2018 to 2021) of physical and chemical properties of $PM_{2.5}$ in Beijing, we observed that primary emissions (e.g., vehicle and cooking activities) in the post-COVID period were further reduced, in addition to the long-term emission control implemented since the early 2010s. Pollutants associated with local primary emissions were suppressed more significantly (e.g. black carbon: -38%), especially during traffic rush hours and cooking time. In the meantime, the contributions of secondary organic aerosol (OA) to primary OA, as indicated by the f_{44}/f_{43} ratio determined by the Time-of-Flight Aerosol Chemical Speciation Monitor, were amplified extensively, especially during heavily polluted days ($PM_{2.5}$ >150 μg/m^3, f_{44}/f_{43}: 1.7 and 2.5 before and after COVID lockdown). Similar reductions in primary emissions during the post-lockdown period, as indicated by the black carbon concentration, were observed in Milan, Italy. Evident decreases in the Chinese megacities (up to 20%) in $PM_{2.5}$-related mortality were also determined using the BENMAP-CE model.

High Number Concentration of Atmospheric Sub-3 nm Particles in Polluted Environment of East China: three years' Observation at SORPES Station

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Atmospheric aerosols are of great concern due to their impacts on human health, visibility, and climate. Sub-3 nanometer particles are initially formed through gas-to-particle conversion with a typical size of 1-3 nm, which is crucial for understanding the steps of new particle formation and sources of aerosol particles in the atmosphere. In this study, sub-3 nm particle size distribution was measured by a Particle Size Magnifier (PSM) from 2018 to 2020 at Station for Observing Regional Processes of the Earth System (SORPES), China. Based on three years' measurement, we present observations of a high concentration of sub-3 nm particles in eastern China, with magnitudes ranging from 10^3 to 10^5 cm^{-3}, which is approximately two orders of magnitude higher than that observed in the boreal forest environment.

We found that secondary formation is the dominant source of sub-3 nm particles during the daytime. The overall occurrence frequency of new particle formation (NPF) events was found to be over 40%, with higher formation rates driven by the high concentration of sulfuric acid (SA) in polluted areas. In addition, primary emission from heavy traffic was an important source of sub-3 nm particles in urban areas. To quantify the contributions of these sources, we used SA, NOx, and condensation sink as tracers. Our analysis revealed that during the daytime, traffic emissions can contribute over 50% to sub-3 nm particle concentrations on non-NPF event days, and even up to 13% on NPF event days. During the nighttime, the dominant contribution of traffic emissions can be up to 70%. Furthermore, synergistic effects of traffic emissions and new particle formation can occur simultaneously during the daytime, leading to extremely high concentrations of sub-3 nm particles. Our study emphasizes the importance of considering traffic emissions as a primary source of sub-3 nm particles in anthropogenic pollution areas in regional and global air quality models.

Reduced aerosol pollution and intensified summertime rainfall under COVID-19 lockdown in India

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Understanding impacts of aerosol loading on precipitation is an important and urgent need for water resource management and disaster prevention in India. In this study, impacts of COVID-19 emission reductions on precipitation are investigated. We focus on the record-break rainfall enhancement in May 2021 over whole India when second lockdown occurred. Future shift of precipitation in India under a lower aerosol emission condition is also evaluated by using the Coupled Model Intercomparison Project Phase 6 (CMIP6) model outputs. We find that emission reductions during the lockdown period enhance moisture transport from ocean to land and intensify vertical atmospheric convection over India. Future model simulations also suggest an increasing trend in monsoon precipitation under low emission condition.

Biomass burning organic aerosol: phase separation and viscosity

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Worsening air quality is a global concern but is particularly important in developing countries and large cities where pollution levels often surpass recommended limits. Emissions from biomass burning are among the largest global contributors to organic aerosol, significantly impacting human health, air pollution and the climate (1). The air quality in urban environments is heavily influenced by biomass burning, from both rural and urban sources like agricultural waste burns, forest fires, and residential burning.

An improved understanding of the phase behavior and viscosity of biomass burning organic aerosol (BBOA) is needed to understand its environmental impact. Viscosity can affect aerosol growth rates, size distributions and heterogeneous reaction rates (2), including the whitening rate of brown carbon in BBOA that ultimately affects the Earth's radiative budget. The phase behavior (how many phases coexist in particles and their morphology) is important for the equilibrium partitioning of semi-volatile organic compounds, reaction rates and influencing cloud nucleation ability of BBOA (3).

We study lab-generated BBOA formed from wood smoldering in a controlled furnace tube. We observe the phase behavior of BBOA particles directly sampled from the woodsmoke, and identify that BBOA from wood smoldering undergoes liquid-liquid phase separation into core-shell particles. Fluorescence recovery after photobleaching (FRAP) experiments allow calculations of the viscosity of the different BBOA phases (4). We show that the organic shell on the outside of a BBOA particle exhibits viscosities that are, under certain tropospheric conditions, orders of magnitude higher than that of the more-polar core. The viscous organic shell could present kinetic limitations towards gas and VOC partitioning and diffusion within particles, with important implications for potentially slowing heterogeneous reaction rates and extending lifetimes of toxic pollutants.

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Molecular characterization of organic aerosols in two heavily polluted cities of India: seasonality and sources

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Fine particulate matter ($PM_{2.5}$) poses a major threat to air quality, climate, and public health. India is experiencing severe PM pollution, with a substantial contribution from organic aerosols (OA) [1]. Previous observations typically focused on low chemical resolution analysis within short period, hindering the understanding on OA sources/formation and seasonality [2]. To address those knowledge gaps, we conducted a year-long period offline campaign of $PM_{2.5}$ at two most polluted cities in India, New Delhi, and Kanpur. The water-soluble organic compositions were analyzed with three advanced mass spectrometers, i.e., an Aerodyne Aerosol Long Time of Flight Mass Spectrometer (LTOF-AMS), an extractive electrospray ionization (EESI)-LTOF with soft ionization strategy [3], and a combination of EESI and ultrahigh-resolution Orbitrap mass analyzer for near molecular aerosols speciation.

AMS-based Positive matrix factorization (PMF) identified 2 primary and 2 secondary factors. The two dominant oxygenated factor accounted for 71.1% in Kanpur and 61.2% in New Delhi over the year. The primary OA affected by traffic and solid fossil fuels contributed more on Delhi while biomass burning OA (BBOA) had a higher contribution in Kanpur. The near-molecular EESI PMF analysis further identified 7 factors, including winter primary/secondary OA, nitrogen-containing OA, cooking OA, post-monsoon OA, cigarette/incense burning OA and BBOA. Furthermore, Lagrangian plume tracking combined with fire spots maps further attributed the post-monsoon factor to significant influences from agricultural residues burning, particularly in Punjab during October and November. The source apportionment insights gained from this work will help in better identification of key OA sources and secondary processes in India.

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The Frequency of Sand and Dust Storms in the Eastern Mediterranean Region and its Possible Health Risk

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High concentrations of airborne dust on top of the atmosphere have a substantial impact on radiative forces, energy sector, and human health [1–3]. In fact, exposure to airborne particulate matter during a sand and dust storm (SDS) stimulates serious health effects, especially for the vulnerable groups. Dust particles may include trace metals, fluoride, natural asbestiform compounds, radioactive elements, silicates, alkali salts, and bio allergens.

Quantify the amount of deposited dust particles in the respiratory tracts is the first step in the process to understand the effects of such exposure. Here, a simple model was utilized to quantify that based on a long-term observation in an urban background site in an Eastern Mediterranean city (Amman, Jordan) during 2013–2019. This approach was applied adult males/females during identified SDS. Several human activities were considered in the exposure scenarios (resting and exercising).

During a typical SDS episode, the dust particle number size distribution can be characterized by two main modes: accumulation and coarse that have geometric mean diameters of about 0.5 μm and 2.5 μm; respectively. The corresponding particle number concentration can be in the range 100–1000 cm^{-3} and 10–100 cm^{-3}; respectively. Assuming the bulk density of dust particles about 2.6 g/cm^{-3}, this yields total dose rate for an adult male 3–15 mg/h and 28–154 mg/h during the minimum exposure level and the maximum exposure level; respectively.

The importance of this investigation relies on the fact that the Eastern Mediterranean region has been affected by SDS episodes more frequently in the past decades as a result of climate change, desertification, and war [4–8].

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Oxidation Products and Aerosol Yields from D5 Siloxane, an Emerging Urban Pollutant

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Siloxanes are anthropogenic pollutants emitted from consumer and industrial products and are constituted of silicon, oxygen, and alkyl groups. Decamethylcyclopentasiloxane (D_5 siloxane) is a ubiquitous cyclic siloxane in human environments, and recent smog chamber and oxidation flow reactor experiments have found that D_5+OH would form secondary organosiloxane aerosol (SOSA).

However, there is uncertainty about the volatile organic products (VOP), and there are wide discrepancies in the reported aerosol mass yields (Y_{SOSA}). To identify the VOP and to reconcile Y_{SOSA}, we performed experiments with varying D_5 concentrations and OH exposures in a Potential Aerosol Mass oxidation flow reactor with 185 nm UV lamps. We used a proton transfer reaction time-of-flight mass spectrometer and a scanning mobility particle sizer to detect VOP and to quantify Y_{SOSA}.

We found molar yields of HCHO and HCOOH at >100 % and >40 % respectively at ~1 day of OH exposure. Larger VOP, tentatively identified as silanols and formate esters, were also detected. As for the aerosol, we found Y_{SOSA} to be small (<10 %) at ~1 day of OH exposure and ~20 % at the OH exposure corresponding to the atmospheric lifetime of D_5. Combining the data from these experiments and those in the literature, we found that a volatility basis set model incorporating both OA mass loadings and OH exposure would adequately parameterize Y_{SOSA}.

Elucidating Variations in Molecular-Level Speciation and Sources of Ambient Organic Aerosol in Two Megacities of North and South India

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Keywords: source apportionment, extractive electrospray mass spectrometry, water-soluble mass
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Air pollution control strategies in India suffer from knowledge gaps in chemical speciation of $PM_{2.5}$, which is needed for isolating individual source contributions. We collected $PM_{2.5}$ filter samples from four centrally located sites in two select megacities of India – Lucknow and Pune, between Dec 2020 and May 2022 spanning both winter and summer seasons. Lucknow is located in the Indo-Gangetic plains of the north while Pune is part of the Deccan plateau region in the south. The water-soluble organic mass (WSOM) of filter samples was analyzed using a time-of-flight aerosol mass spectrometer (ToF-AMS), extractive electrospray ionization time-of-flight (EESI-ToF) MS and total organic carbon analyzer. The elemental composition, inorganic ions and organic tracers for secondary organic aerosol were also measured. Subsequently, source apportionment analysis was performed using the Source Finder software to identify major sources and their relative contributions.

The maximum wintertime WSOM concentration in Lucknow (~120 µg m^{-3}) reached 5 times that in Pune (22 µg m^{-3}) before reducing to 15 – 30 µg m^{-3} at the onset of summer in April, which still exceeded summertime Pune concentrations (4 – 8 µg m^{-3}). The water-soluble organic carbon constituted 35 – 40% of the total organic carbon in these samples suggesting that total organic aerosol likely exceeded 200 µg m^{-3} in Lucknow and 50 µg m^{-3} in Pune. Source apportionment results revealed that wintertime WSOM in Lucknow was dominated by local and regional-scale biomass burning emissions that are typical in the Indo-Gangetic plains along with sulfur-containing emissions. Biomass-burning OA showed aged mass spectral features in Pune suggesting mainly regional-scale influence. At the molecular level, $C_xH_yO_z$ and $C_xH_yN_pO_z$ species constituted the majority of WSOM composition with C# and O# spanning from 5-16 and 1-6 respectively. Interestingly, factors with similar ToF-AMS spectral features between the two cities showed differences in molecular-level factor profiles obtained from source apportionment of EESI-ToF measurements indicating source-type differences which could be isolated mainly via soft ionization-based EESI-ToF measurements. These results help identify sub-sectoral air pollution sources based on molecular-level speciation and temporal trends that would be useful in formulating effective air pollution control policies to protect public health.

Numerical simulation and evaluation of global ultrafine particle concentrations

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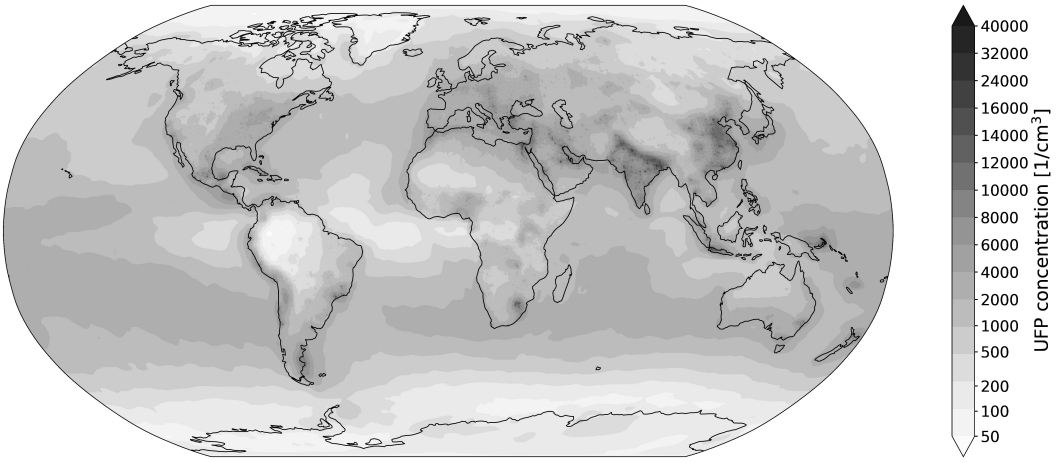
Ultrafine particles (UFP), i.e. particles with an aerodynamic diameter below 100 nm, have a significant impact on public health and the hydrological cycle. Recent studies showed that their ability to penetrate more deeply into the lungs and potentially into the blood stream may cause an increased incidence of cardiovascular and cerebrovascular diseases. Additionally, UFPs significantly contribute to cloud condensation nuclei concentrations. However, knowledge on global distributions of UFPs is scarce.

We present a global simulation of UFP concentrations using the ECHAM/MESSy Atmospheric Chemistry Model (EMAC), including tropospheric and middle-atmospheric processes, and the modal aerosol microphysics submodel GMXe. Model results were evaluated over Europe, the United States, India and China, using particle size distribution and particle number concentration measurements from available datasets and the literature. An observation-guided downscaling based on high-resolution anthropogenic emission datasets yields UFP concentrations with unprecedented 0.1 x 0.1° horizontal resolution at the Earth's surface, improving the agreement with observations, decreasing the root mean squared logarithmic error and removing discrepancies associated with air quality and population density gradients within the relatively large model grid boxes. We show the importance of primary emissions and nucleation on UFP concentrations as well as their composition and seasonality.

Furthermore, the model setup is extended to additionally represent aerosol processes in the free troposphere and the UTLS region, as a basis for studies on the abundance, composition, formation, transport mechanisms and optical properties of aerosols in the middle atmosphere. We present an outlook and first results of the ongoing studies.



The *Kinetic Laboratory Compass*: Using kinetic models and machine learning for experiment design

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Multiphase chemical kinetics models are commonly applied and successful tools in science and engineering with use cases in, e.g., atmospheric, physiological and technical chemistry, reactor design or process optimization. The associated chemical, physical or biological systems are often influenced by a large number of kinetic parameters, such as reaction, diffusion or partitioning coefficients. Due to their coupled nature, constraining these parameters with experimental data can be difficult and experimenters have to rely on instinct and experience to choose experimental conditions that allow to extract enough information about the system (Berkemeier et al. 2021). We present the *Kinetic Laboratory Compass*, a quantitative tool for experiment prioritization coupling kinetic multi-layer models, global optimization, ensemble methods and optionally machine learning. We demonstrate the method by evaluating the outputs of the kinetic multi-layer model of aerosol surface and bulk chemistry (KM-SUB, Shiraiwa et al. 2010) under conditions that represent practicable laboratory experiments, addressing the ozonolysis of oleic acid aerosols. For the quantification of constraint potentials associated with these potential experiments, we developed two metrics, the *Ensemble Spread* and *Parameter Constraint Potential*. The methods are tested in simulations that include experiment selection, conduction and global optimization. We find that the demonstrated techniques achieve much larger constraints of the model solution space or of individual kinetic parameters, as randomly selected experiments. Using an artificial neural network surrogate model (Berkemeier et al. 2022), the process can be significantly accelerated with little dissimilarity of the results in comparison with the template model application. The presented workflow is adaptable to various process models to streamline the costly process of experimental validation and optimization. Furthermore, our findings emphasize the applicability of machine learning surrogate models for computationally expensive process models and applications.

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Investigation of the development of global aerosol regimes from pre-industrial times to the future based on global aerosol simulations and machine learning techniques

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The temporal development of the global distribution of aerosol regimes, i.e. areas of specific aerosol characteristics, is investigated based on global aerosol simulations performed for six time slices, including pre-industrial (1750, 1850) and present-day conditions (2015), as well as future (2050) following the Shared Socioeconomic Pathways SSP3-7.0, SSP2-4.5 and SSP1-1.9. The aerosol characteristics are evaluated in terms of aerosol regimes by combining the information about multiple aerosol properties within a cluster analysis. The present-day case serves as a reference where aerosol regimes are identified via the unsupervised machine learning technique K-means. In addition, a supervised machine learning technique (Random Forest) is applied on the basis of the aerosol regime classification criteria gained from the reference case. This allows for a consistent identification of comparable aerosol regimes across the different time slices. With this approach we analyze, for instance, which present-day aerosol regimes could have been present in the past and how they might develop in the future. These analyses are conducted for three atmospheric altitude layers with a special focus on the lower troposphere. After conducting a primary classification, we also implement a secondary sub-classification to assess the fine structure of the identified primary regimes. The results are further complemented by an in-depth analysis of the emissions of primary aerosol species and aerosol precursor gases for the different time slices in selected regions, including information about the respective contributions from different emission sectors. Details will be presented on a poster.

Enigma of biogenic VOCs in highland ecosystem: characteristics, emissions and sources based on in-situ observations at the Qinghai-Tibet Plateau

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Oxidation of volatile organic compounds (VOCs) is one of the main causes of the formation of secondary pollutants, such as SOA, with biogenic VOCs (BVOCs) playing a critical role due to high emission rate and reactivity. The Qinghai-Tibetan Plateau is an exceptional platform, but the lack of ground-based measurements limits our capacity to investigate BVOCs concentrations and their atmospheric effects.

The vegetation of the Qinghai-Tibetan Plateau is mainly composed of alpine meadows. We conducted in-situ observations at four sites using PTR-ToF 1000. Variations between sites were observed, with high abiogenic isoprene (IP) at Golmud. Lulang with abundant coniferous forests, results in significant monoterpenes (MTs). Both the Lulang and the Magu sites were within the NOx control area. The MTs/IP ratio at Lulang was higher than 1:1, while the others were around 1:3 or less.

The air trajectories at Lulang reveals the influence of the westerly wind band, whereas a minor part was affected by South Asian monsoon. Aside from large-scale atmospheric circulation, the unique mountain-valley breeze can also have a substantial impact. BVOCs is impacted by a combination of local emissions superimposed on transport. MTs were regulated by a combination of humidity and temperature changes. The Model of Emissions of Gases and Aerosols from Nature (MEGAN) was used to estimate the BVOCs emission rates at Lulang and the MT was found to be highest, reaching 5E-7 kg C/(m^2h). Using a back-calculation to estimate the true emission concentrations based on observation. Then 42% of the BVOCs was consumed and only 58% of the original emissions were captured. Upon comparing the observed and simulated emission rates, a good agreement was achieved, with MEGAN being approximately 3-6 times higher.

In-situ observations in the Qinghai-Tibetan Plateau have furnished evidence for the characterization, variability, and sources, which are in good agreement with simulation. In order to gain a better understanding of the causes and mechanisms of air pollution and to formulate mitigation policies, more observations and analyses supplemented by additional data sources (e.g. OOMs) are needed in future studies.

Importance of Bias Correction in Data Assimilation of Multiple Observations Over Eastern China Using WRF-Chem/DART

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Three types of observations, aerosol optical depth from the Moderate Resolution Imaging Spectroradiometer, surface particulate matter with diameters less than 2.5 ($PM_{2.5}$) and 10 μm (PM_{10}), and aerosol extinction coefficient (AEXT) profiles from ground‐based lidars, were separately and simultaneously assimilated using the Weather Research and Forecasting Model with the Chemistry/Data Assimilation Research Testbed (WRF-Chem/DART). Two cases in June and November 2018 were selected over middle and eastern China. Experiments assimilating single-type and multiple observations were evaluated by cross validating their analysis and forecast against the three observation types. Compared to the experiment without data assimilation (DA), DA of single-type observations is always closer to the type of observations assimilated. However, DA of aerosol optical depth or AEXT sometimes significantly degraded the error performance for $PM_{2.5}$. This problem is caused by the inconsistency of bias tendencies when modeling aerosol optical properties and surface aerosol mass. It is found that WRF-Chem tends to predict dryer air within the boundary layer over eastern China, which may have played a role in the underestimation of AEXT even when $PM_{2.5}$ was overestimated. After applying a simple bias correction (BC), the problem was alleviated. DA of multiple observations with BC gives the best overall error performance when validated against all types of observations and even performs better than any DA of single-type observations experiments in reproducing AEXT profiles. The results illustrate that BC is important in DA of multiple observations and that the simultaneous DA of aerosol observations with different vertical information can work synergistically to improve aerosol forecasts.

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A new airborne aerosol sampling system: development, validation, and application in vertical profile measurements of aerosol properties

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Vertical profile measurements of aerosol physicochemical properties play a key role in addressing aerosol environment and climate effects. Traditional in-situ vertical observations of those properties are mainly based on aircraft platforms, which are costly and restrictive, and not applicable for near-ground (<500 m) measurements due to safety reasons. Tethered balloon (TB) and unmanned aerial vehicle (UAV) are ideal observation platforms for vertical measurements within 1-km height but only suitable for small portable instruments due to their payload limitations (typically < 10 kg). In this study, a new lightweight aerosol sampling system is developed for TB and UAV platform. The system can collect aerosol samples at up to 12 heights and store them at aerosol state in conductive bags. The samples (with a typical volume of 3 L/sample) can be directly analyzed by most of online aerosol instruments such as aerosol mass spectrometer, volatility/hygroscopicity tandem differential mobility analyzer (V/H-TDMA), and single particle soot photometer (SP2), etc. The new system provides an easy and economical way for the vertical profile measurements of aerosol physicochemical properties. During three intensive field campaigns conducted in the Qinghai-Tibet plateau, this new system combined with a SP2 and a HTDMA was first applied to determine the vertical profiles of refractory black carbon (rBC) mixing state and aerosol hygroscopicity. Results show that most rBC-containing particles are externally mixed and the proportion of internally mixed rBC increases with height. The hygroscopicity of aerosol particles in accumulation mode peaks at the height of 100-200 m during night and morning time. The vertical profiles of aerosol hygroscopicity and rBC mixing state show clear diurnal variations, driven by surface emissions, aging processes, and evolution of boundary layer.

Number of phases in organic aerosol mixtures is driven by difference in oxygen-to-carbon ratio of components

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Atmospheric aerosol particles play an important role for air quality and climate. Primary organic aerosol (POA) and secondary organic aerosol (SOA) make up a significant mass fraction of these particles. POA denotes aerosols that are directly emitted into the atmosphere as particles, while SOA mostly forms from oxidation of gases, followed by gas-particle conversion of the oxidation products. Representation of SOA formation in models and prediction of their impact on air quality and climate requires knowledge of their phase behavior (the number and types of phases) in internal mixtures of POA and SOA. For instance, it is often assumed that SOA formation is enhanced in the presence of POA particles due to a lowering of the activities in the organic aerosol phase, and hence a lowering of the equilibrium partial pressure, in case of single-phase POA+SOA particles. The presence of POA will have a smaller effect on the formation of SOA mass in cases of phase-separated particles.

Here, using microscopy, we observed the phase behavior of individual particles containing mixtures of POA and SOA. Eleven different, commercially available organic molecules were used as proxies for POA, covering a range of oxygen-to-carbon (O/C) ratios between 0 and 0.29. Each of the hydrocarbon-like POA proxies was internally mixed with one out of seven different SOA materials generated in environmental chambers. The majority (88%) of the mixtures resulted in phase-separated particles, and the phase behavior was found to be independent of the relative humidity between 90% to 0%. We show that the difference in the average O/C ratio (a proxy for difference in polarity) between the POA and SOA component of a mixture is a good predictor of the phase behavior with two-phase particles forming for $\Delta O/C \geq 0.265$. Our results have important implications for air pollution policies being considered to limit SOA formation in urban environments.

Source contributions and drivers of physiological and psychophysical co-benefits from major air pollution control actions in North China

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North China is among the most polluted regions in the country, and human exposure to $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5 μm) in this region has led to severe health consequences. The region has also benefited the most from emission reductions in recent years. It is of interest to understand to what extent and through which paths emissions from different sectors cause adverse health impacts. Here, we present the results of a full evaluation of the health benefits of emission control actions implemented in recent years based on segregated emission inventories with an emphasis on residential emissions. Two major causal paths, one from residential emissions to indoor air pollution, exposure, and premature deaths, and the other from non-residential emissions to ambient air pollution and psychophysical impacts, were identified and quantified. From 2014 to 2019, both ambient (33%) and indoor (39%) $PM_{2.5}$ decreased significantly, leading to decreasing trends in exposure (36%), premature deaths (10%), and psychophysical impacts (21%). The Air Pollution Prevention and Control Action Plan, the Clean Heating Campaign and spontaneous residential shifts to clean energy contributed significantly to these reductions when the effects of other drivers, such as population and economic growth, were excluded.

MultilayerPy: a tool for creating and optimising multi-layer models of aerosol and film processes

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Heterogeneous processes such as aerosol-gas chemical reactions and vapour uptake are key to understanding the behaviour of aerosols in our environment. They contribute to their ability to take up water to form cloud droplets and determine the persistence of harmful particle-bound compounds, impacting the climate and human health.

Kinetic multi-layer models such as the kinetic multi-layer model for aerosol surface and bulk chemistry (KM-SUB)[1] and gas-particle interactions (KM-GAP)[2] are state-of-the-art models used to describe these processes on the particle and film level. KM-SUB and KM-GAP-based models have been used to determine the oxidative potential of particulate matter, the impact of surfactant self-organisation on aerosol chemical lifetimes, and the impact of aerosol phase state on the long-range transport of toxic chemicals. These models are useful but cumbersome to write and there is a need for an open-source tool to assist researchers in creating and optimising them.

We have developed MultilayerPy, an open-source Python package which facilitates the creation and optimisation of kinetic multi-layer models.[3] This software is written such that the user uses building blocks (i.e. reaction scheme, bulk diffusion parameterisations, and model components) to automatically generate model code which can then be ran and the output presented in a reproducible manner. This reduces the time needed to develop model descriptions of aerosol processes and allows the user to focus on the scientific issues rather than coding the models. I will present recent use cases of the software looking at the chemical lifetime of real aerosol material in the atmosphere, along with ongoing work extending the base package.

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The effect of biological and atmospheric oxidants on proteins in the epithelial lining fluid

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Biological systems are subject to oxidants. Radical production is part of normal cellular regulation and essential for cell homeostasis, however, enhanced exposure to oxidants may result in an imbalance of oxidants and antioxidants in the body and lead to oxidative stress [1].

Exposure to ambient air pollution can cause adverse health outcomes and mortality. [2] Pollutants including fine particulate matter with a diameter less than 2.5 µm (PM2.5) and gaseous oxidants such as ozone (O3) and nitrogen oxides (NOx=NO+NO2) are the most toxic components of pollution.

Among all biological molecules, proteins are the main target of oxygen- and nitrogen-derived reactive species because of their rapid reaction rates with oxidants and their high abundance in the cells and extracellular fluids, such as the epithelial lining fluid [3]. The kinetics of the interactions of gaseous air pollutants, PM2.5 and proteins in the lung have not been studied in detail previously. Here, we evaluate the damage to proteins by biological and atmospheric oxidants using a kinetic model of the human respiratory tract, KM-SUB-LUNG, and attempt to link our model results to measurement biomarkers of oxidative stress. Our findings suggest that O3 and NO2 are a large burden on the antioxidant system, but often do not lead to the formation of biomarkers associated with oxidative stress. Exposure with PM2.5 leads to formation of highly reactive OH radicals and dominates the production of common markers such as nitro-tyrosine.

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Effects of long-term ambient fine particulate matter exposure on asthma: Evidence for children and adults

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The effects of long-term fine particulate matter ($PM_{2.5}$) exposure on asthma, especially in adults, remains inconclusive. Benefiting from the recent explosion of relevant epidemiological studies, we comprehensively explored the effects of long-term ambient $PM_{2.5}$ exposure on both childhood and adult asthma in light of the emerging evidence by combining multiple state-of-the-art approaches.

First, we explored the association between long-term $PM_{2.5}$ exposure and risk of asthma by conducting a systematic review and meta-analysis. We concluded that the association is statistically significantly positive both in children and adults. We then estimated the exposure-response curves for asthma—i.e., explored the exposure-response effect of asthma at different exposure levels, by constructing exposure-response functions (ERFs) based on data extracted from the systematic review. We find that risk of childhood asthma increases almost linearly with $PM_{2.5}$ concentrations, while the risk of adult asthma increases sub-linearly. In addition, the risk of childhood asthma is much higher than that of adult asthma at any given exposure level.

After confirming by the meta-analysis and exposure-response analysis that $PM_{2.5}$ exposure is statistically significantly associated with increased risk of asthma, we estimated the global burden of asthma attributable to $PM_{2.5}$ by applying the ERFs in an epidemiological model. We find that $PM_{2.5}$ exposure is responsible for 2.9 (95% CI: 1.7–3.9) million years of life lost (YLLs) and 2.3 (95% CI: 1.5–2.9) million years of healthy life lost due to disability (YLDs) through asthma, indicating large adverse effects on public health—i.e., the overall disease burden caused by $PM_{2.5}$ is much higher than previously thought, and therefore more stringent legislations are required to improve air quality as soon as possible.

Our study provides additional evidence on the effects of long-term $PM_{2.5}$ exposure on asthma and calls for greater attention to the effects as they contribute to substantial disease burden. In addition, the exposure-response curves established in our study—which incorporating evidence on high exposure levels that covering most of the worldwide exposure ranges—could be applied to assess the city to global scale asthma-related health benefits obtained from air pollutant reduction associated with policy scenarios.

EURO 6 compliant gasoline vehicle emissions and SOA formation after aging in the photochemical emission aging flow tube reactor during steady state driving

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To improve air quality, EU guidelines for Light-duty vehicle emissions are frequently updated to use and force new emission reduction technology. The Euro 6 emission standard introduced a limit to the particle number concentration of $6.0 \cdot 10^{11}\ \#/km$ emitted by Gasoline Direct Injection (GDI) vehicles. Thus, gasoline particle filters (GPF) were introduced to modern vehicles to lower particles' emissions. However, current regulations do not limit the formation of secondary organic aerosols (SOA) from Volatile Organic Compounds (VOCs) emitted. Within the Helmholtz International Lab aeroHEALTH, emissions from the exhaust of a Euro 6 standard-compliant car were studied. The car operated under 50, 80 and 100 km/h, and idling. The exhaust was aged in the Photochemical Emission Aging Flow tube Reactor (PEAR), simulating 2 days of atmospheric equivalent aging by reactions with OH radicals. Both gas and aerosol phase were comprehensively characterized before and after aging in the PEAR.

The emissions of primary particles (diameter > 10 nm) were less than 7 #/m³. However, after aging, SOA concentrations increased significantly and ranged between 1.5mg/m³ and 10.5 mg/m³ depending on the driving conditions. The SOA mass by distance driven was 0.5-0.8 mg/km at 80-100 km/h and 1.0-1.2 mg/km at 50 km/h. At 50 km/h the observed SOA formation is comparable to recent studies for euro 3-5 vehicles. Based on literature SOA yields, up to 50 % of the total SOA concentration after aging in the PEAR could be explained by the three most abundant non-benzene aromatic compounds (toluene, xylene, and trimethyl-benzene) present in the exhaust emissions. Further, long Idling phases (> 5 min) increased the SOA concentrations from 0.7 mg/m³ to 6.5 mg/m³. The study demonstrates that the GPF addition successfully reduces primary particle emissions. However, SOA formation after aging remains an issue that degrades air quality and can pose a significant health risk in the urban environment.

Glyoxal as a Potential Source of Highly Viscous Aerosol Particles

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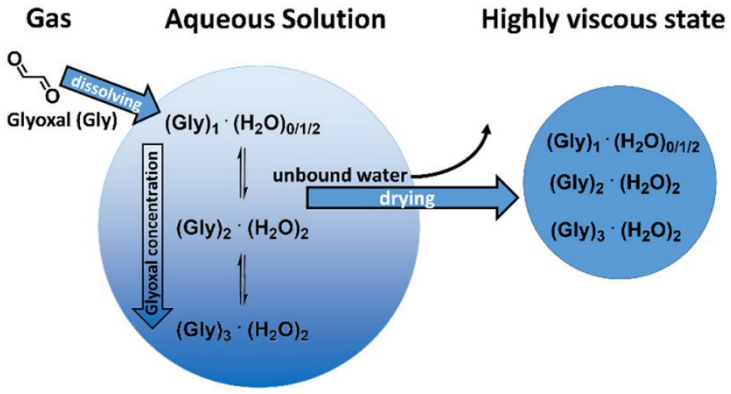
Recent studies have shown that glyoxal may remain in the particle phase of aqueous aerosol particles upon drying despite the high vapor pressure of pure glyoxal, due to the formation of oligomeric glyoxal water adducts with low vapor pressure. Little is known about the phase state of such particles, even though some studies suggested a semisolid or glassy state for dried aqueous glyoxal solutions. For the study presented on this poster, we performed glass transition temperature (T_g) measurements on various aqueous glyoxal systems. We show experimentally that very slow and also fast drying of aqueous glyoxal solutions can indeed lead to the formation of highly viscous semisolid and glassy states, both in bulk as well as in aerosolized samples. T_g changes with the solute concentration before drying, with drying rate and in the presence of additional solutes such as ammonium sulfate or ammonium bisulfate, even when they are present only in catalytic amounts. Temperature-dependent measurements show that the equilibration between various glyoxal species upon water addition, mimicking atmospheric water uptake upon rising humidity, can range from hours to days. We use the measured glass transition temperatures to infer dependencies of the aqueous phase equilibria between monomer, dimer, and trimer glyoxal species and their water adducts and support these by infrared spectroscopy. Our results imply that aqueous glyoxal aerosols may form highly viscous states at atmospherically relevant conditions.[1]



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The influence of different types of SOA on climate and human health

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Atmospheric aerosols significantly affect visibility, air quality and climate. Exposure to atmospheric aerosols has been associated with increased cardiopulmonary mortality and morbidity. Directly emitted hydrocarbon components are referred to as primary organic aerosol (POA) and secondary organic aerosols (SOA) can be formed via gas-phase reactions and the subsequent condensation of semi-volatile vapour, as well as by multiphase and heterogeneous processes.

SOA accounts for as much as 50 – 90 % of the total organic aerosol (OA) burden and has recently received much attention. Moreover, SOA also contributes importantly to the Earth's radiation balance through its absorption and scattering of solar radiation and by altering cloud microphysical properties since it contains not only colorless carbon, which merely scatters light, but also colored carbon, which can absorb light.

However, the quantification of SOA sources and/or pathways and the climatic effect of SOA are more challenging mostly due to poor understanding and measurement of the SOA species, consisting of thousands of multifunctional, oxygenated species to a highly varing degree and including high molecular weight species and oligomers. Limitations in SOA source apportionment are tied directly to limitations of the measuring instruments nowadays around the world, which either involve thermal decomposition, or have low time and mass resolution. In consequence, the exact SOA sources and PM chemical composition along with its mass concentrations at molecular level for predicting the overall health impacts associated with aerosol exposure remain unknown at present.

Here, we explore the SOA sources of organic aerosol, by combining field observations and air-transport modelling, and find that the different pollution events are controlled by different SOA sources. For this, we have elucidate the chemical composition at molecular level and predominant SOA sources during different pollution episodes by combining the most advanced instrumentation with Ultra-high mass resolution (EESI-Orbitrap) as the first field application and comprehensive analytical approaches. In addition, an Extractive electrospray ionization long time-of-flight mass spectrometer (EESI-LTOF), an HR-AMS (Aerodyne Research Inc.), and ROS, were also deployed.

Measurements were performed from 26 January to 20 February 2021 (covered the Chinese New Year, CNY) at the Tower Site of the Institute of Atmospheric Physics (IAP). The site is located between the third ring and the forth ring roads in northern Beijing, surrounded by residential infrastructure and is characterized as an urban background site.

Furthermore, we use field observations to derive the brown carbon optical properties and OP parameters impacted by SOA sources. Our results demonstrate the necessity of the characterization of SOA sources and its implication on climate and human health. Note that Beijing, China experiences severe air pollution events in winter with complex primary emissions and secondary formation mechanisms, so it was chose to be the most suitable site for the investigation of SOA sources. The further detailed results will be shown at the conference.

Heterogeneous chemistry and nanometer particle growth: laboratory studies on transesterification in nanometer-sized aerosol particles

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The interest in researching new aerosol particle formation and their growth has been gaining. After the initial formation via nucleation of nanometer-sized aerosol particles, they grow governed by organic molecules. Especially the small particle embryos offer a unique nanoscale chemical environment – due to their size-dependent physical properties, they might influence chemical reactions within the particle phase. One consideration is the higher effect of interface curvature present in smaller particles, which could impact the reaction rates and the equilibrium status of chemical reactions. For example, condensation reactions including the elimination of highly volatile molecules should be favored. The volatilization of the more volatile product out of the condensed phase should be enhanced, favoring the formation of higher molecular weight products. Thus, particle size-dependent chemical reactions could play a crucial role in the life cycle of atmospheric aerosols. In our studies, we plan on focusing our research on investigating transesterification reactions as representatives of condensation reactions, using methyl-esters to generate methanol as a highly volatile product. However, the aerosol particles' growth and behavior, alongside product formation, will be examined. For analysis, Scanning Mobility Particle Sizer Systems (SMPS) and a GC-MS system with thermal desorption will be utilized.

Satellite proxies for estimating spatial variation of new particle formation

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In situ measurements are the only observational method to obtain information on new particle formation (NPF) events and particle growth. However, they are bound to a certain location and often lack the information on regional scale variability. Satellites have the advantage of providing global observations on multiple atmospheric components but freshly formed, nano-sized aerosol particles are far too small to be observed with optical remote sensing measurements. Satellite instruments can observe aerosols only after they have grown to optically active sizes (diameter > about 0.1 μm). Even though direct observation of nanoparticles is not possible with satellites, they provide relevant information on nanoparticle precursors, UV- radiation and optically active aerosols. In this work we investigate the capabilities of current operational satellites to provide information on NPF by developing satellite-based proxies that are formulated by source and sink terms for atmospheric nanoparticles. The overall goal is to use the satellite-based proxies to obtain information on gigacity-scale variability of NPF.

The satellite-based approach for obtaining information on NPF is based on a hypothesis that the formation of atmospheric nanoparticles is governed by sink and source terms. The main sink for the nanoparticles and precursor vapours is condensation sink to the pre-existing aerosol particles, that can be estimated using satellite-based aerosol optical depth (AOD), or modified AOD observations. The source term, on the other hand, is related to UV-radiation and precursor gases. With the help of detailed in situ measurements, we are able to derive and test different formulations of satellite-based proxies for NPF.

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Cirrus temperature ice nucleation abilities of soot from commercial aircraft engines

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Aircrafts operate mainly in the upper troposphere, where the background aerosol (number) concentration is low. At these altitudes, cirrus clouds originate from the homogeneous freezing of small liquid droplets and/or from heterogeneous ice nucleation catalyzed by ice nucleating particles (INPs). Soot particles can act as INPs and have been shown to promote cirrus-cloud formation [1]. However, impacts of aviation soot particles on natural cirrus remain uncertain [2]. To date, the ice-nucleating abilities of real aircraft soot have not been quantified. This is largely due to the challenge of sampling particles downstream of real turbine engines. In this work, we quantify the ice nucleation activity of soot particles sampled from commercial aircraft turbine engines at the engine test cell of SR Technics at the Zürich airport.

Exhaust particles from commercial turbofan engines at different thrust levels were sampled into a cloud chamber, where the particles experienced cirrus temperatures (T < -40°C) and relative humidity (RH_{ice} > 100%), allowing observation of their ice nucleation abilities. The ice nucleation abilities of the soot particles were also measured after thermally (350°C) stripping volatile components associated with the particles and after cloud processing (i.e., after undergoing an initial freezing cycle, mimicking contrail processing). Unprocessed and cloud processed aircraft soot generally froze at conditions required for homogeneous freezing of solution droplets, limiting their role as potent INPs in the upper troposphere. Up to 8% of large (e.g., > 200nm) stripped and cloud processed soot froze at RH_{ice} below that required for homogeneous freezing of solution droplets in a mechanism attributed to pore condensation and freezing [3]. However, the atmospheric relevance of such large soot particles showing stripped-like surface is limited [4,5], indicating that aircraft soot particles are poor INPs for cirrus formation.

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The export of African dust across the Atlantic and its impact over the Amazon Basin

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The frequent trans-Atlantic transport of African dust plumes toward the Amazon Basin during its wet season (January – April) not only interrupts the near-pristine atmospheric condition there, but also provides nutrient inputs into the Amazon rainforest associated with dust deposition. In this study, we use the chemical transport model GEOS-Chem to study the export of African mineral dust toward the Amazon Basin and the associated impact over the rainforest during the period of 2013 – 2017. The model results are constrained by multiple datasets obtained from AERONET, MODIS, as well as Cayenne site and the Amazon Tall Tower Observatory (ATTO) site in the Amazon Basin. With an optimized particle mass size distribution (PMSD) for dust aerosol, the model well captures the observed export efficiency of African dust along the trans-Atlantic transport. During the wet season, African dust entering the Amazon Basin has surface concentrations of 5.7 ± 1.3 μg m^{-3} (up to 15 μg m^{-3} in the northeast corner), accounting for 47% ± 5.0% (up to 70%) of total aerosols. The frequency of dust events in the Amazon Basin in the wet season is around 18% on average, with maxima over 60% at the northeast coast. During the dust events, AOD over most of the Amazon Basin is dominated by dust. Observed dust peaks over the Amazon Basin are generally associated with relatively higher African dust emissions (including Sahara and Sahel) and longer lifetime of dust along the trans-Atlantic transport, namely higher export efficiency of African dust across the Atlantic Ocean. Associated with dust deposition, we further estimate annual inputs of 52 ± 8.7, 0.97 ± 0.16 and 21 ± 3.6 mg m^{-2} a^{-1} for iron, phosphorus and magnesium deposited into the Amazon rainforest, respectively, which may well compensate the hydrologic losses of nutrients in the forest ecosystem.

By how much can co-condensation of semi-volatile compounds alter clouds?

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Semi-volatile compounds (organics, nitrate, chloride) are ubiquitous in atmospheric aerosols and usually contribute over 50% to the aerosol mass worldwide[1]. These semi-volatiles can co-condense with water vapour, which enhances aerosol hygroscopic growth and facilitates their activation to cloud droplets thus affecting cloud formation and Earth radiation balance. Yet, this co-condensation effect is not well constrained as the loss of semi-volatiles during drying and heating in traditional instruments is poorly understood (e.g., HTDMA, CCN counter). To solve this, we developed a novel method to estimate aerosol hygroscopic growth with considering the co-condensation effect from open-access data (visibility, PM2.5 and meteorology)[2]. By applying it in Delhi (India), we found that the co-condensation of HCl can double the aerosol light extinction coefficient and halve the critical supersaturation needed for cloud droplet activation[3]. A similar significant co-condensation effect is found in Chinese megacities (Beijing, Guangzhou, and Shanghai), but with HNO3.

The next question is how significant the co-condensation effect is over a global scale and how much it alters clouds and radiation. Here, we combine novel field observation, a cloud parcel model, and an aerosol-climate model to disentangle this question. Our climate model sensitivity study showed that doubling hygroscopicity (representing co-condensation effect) plays a minor role in global average cloud properties, but significantly increases cloud droplet number and liquid cloud cover in polluted regions, e.g. East/Southeast Asia, India, Europe, East US. Consistently, parcel model calculations confirm the enhancement of cloud droplet numbers up to 25% due to co-condensation, depending on the particle and gas composition, particle size distribution, and air parcel cooling rate. Our study will help to develop a cloud activation scheme including the co-condensation effect for aerosol-climate models.

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Budget analysis of Gas-Phase Sulfuric Acid Concentration and its contribution to sulfate in Polluted Yangtze River Delta, East China

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Gaseous sulfuric acid (H_2SO_4) is a crucial precursor for secondary aerosol formation, particularly for new particle formation (NPF) that plays an essential role in the global number budget of aerosol particles and cloud condensation nuclei. In situ measurements of H_2SO_4 are very rare, largely limiting our understanding of global aerosol formation mechanisms. Previous efforts on building empirical proxies for H_2SO_4, mainly derived based on short-term intensive campaigns, have been demonstrated to be not broadly applicable. In this work, we performed comprehensive measurements of H_2SO_4 and related parameters in the polluted Yangtze River Delta in East China during four seasons and developed a physical proxy based on the budget analysis of gaseous H_2SO_4. Besides the photo-oxidation of SO_2, we found that primary emissions can contribute considerably, particularly at night. Dry deposition has the potential to be a non-negligible sink, in addition to condensation onto particle surfaces. Compared with the empirical proxies, the physical proxy was believed to have a wide application, e.g., rebuild the long-term variation of H_2SO_4 in various environments. In addition, gaseous H_2SO_4 condensation was found to be a non-negligible pathway for sulphate formation, contributing up to about 20% to sulphate during the daytime; while about 2% at night, which was influenced by aerosol loadings, radiation, humidity, and SO_2 concentration. These findings can shed some insights into improving the understanding and simulation of H_2SO_4, and sulfate, in the current regional and global air quality models and build a new connection between human activities and climate change through anthropogenic H_2SO_4-induced secondary aerosol formation.

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The role of CO_2, ammonia, and organic acids in buffering atmospheric acidity: the distinct contribution in clouds and aerosols

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Acidity is one central parameter in atmospheric multiphase reactions, influencing aerosol formation and its effects on climate, health and ecosystems. Weak acids and bases, mainly CO_2, organic acids and ammonia, are long considered to play a role in regulating atmospheric acidity. However, unlike the strong acids and bases, their importance and influencing mechanisms in a given aerosol or cloud droplet system remain to be clarified. Here, we investigate this issue with new insights provided by recent advances in the field, in particular the multiphase buffer theory *(Ref. 1)*. We show that, in general, aerosol acidity is primarily buffered by ammonia, with a negligible contribution from CO_2 and a potential contribution from organic acids under certain conditions. For fogs, clouds and rains, CO_2, organic acids, and ammonia may all provide certain buffering under higher pH levels (pH>~4). Despite the 10^4~10^7 lower abundance of ammonia and organic weak acids, their buffering effect can still be comparable with that of CO_2. This is due to that the cloud pH is at the very far end of the CO_2 multiphase buffering range. This Perspective highlights the need for more comprehensive field observations under different conditions, and further studies in the interactions among organic acids, acidity and cloud chemistry.

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