

| First name | Last name | Organization | Co-authors | Abstract title | Abstract |
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| Rima | Baalbaki | University of Helsinki | Michael Pikridas, Tuija Jokinen, Tiia Laurila, Lubna Dada, Spyros Bezantakos, Lauri Ahonen, Kimmo Neitola, Anne Maisser, Elie Bimenyimana, Aliko Christodoulou, Florin Unga, Chrysanthos Savvides, Katrianne Lehtipalo, Juha Kangasluoma, George Biskos, Tuukka Petäjä, Veli-Matti Kerminen, Jean Sciare, Markku Kulmala | New particle formation in the Eastern Mediterranean | Atmospheric new particle formation (NPF) is a significant source of global aerosol particle number load and cloud condensation nuclei (CCN) (Merikanto et al., 2009). Therefore, it has been the focus of many research studies in the past 20 years ranging from atmospheric observations to chamber experiments and conceptual and modelling studies. These studies aim to understand the NPF mechanism, its characteristics, how and when NPF takes place and eventually how it affects global climate. The conditions governing NPF have been shown to vary in different environments. In this context and within the framework of the Eastern Mediterranean and the Middle East – Climate and Atmosphere Research Centre (EMME-CARE) project, we performed one-year continuous measurements of aerosol particles down to ~ 1 nm in diameter. The study aimed to examine the frequency, characteristics and drivers of NPF at a background site in Cyprus. In this talk, we will present the main findings of our observations and an outlook for future studies. |
| Angela | Buchholz | University of Eastern Finland | Angela Buchholz, Sneetha Kommula, Liqin Hao, Iida Pullinen, Lejsh Vettikkat, Arttu Ylissirniö, Sami Romakkaniemi, Ari Leskinen, Siegfried Schobesberger, Annele Virtanen | In- and out-of-cloud measurements at SMEAR IV: Pristine conditions vs an aged forest fire plume | Understanding aerosol-cloud interactions is crucial for improving the representation of cloud, precipitation, and aerosol processes in earth system models. Chemical processes inside cloud droplets have the potential to impact the properties of the cloud droplets and the aerosol particles re-emerging after the cloud dissipates. As part of the FORCeS project, we studied these processes at the Puijo SMEAR IV station, a semi-urban measurement station in central Finland, which is frequently “in-cloud” due to its elevation. Its dual inlet system enables the parallel sampling of the dried total particle distribution and the interstitial particles (i.e. remaining particles after removal of cloud droplets). Similar suites of instruments were situated at the in-cloud station and a lower level, “out-of-cloud” station. We analysed the Cloud Condensation Nuclei (CCN) activity before, during, and after a cloud event (i.e., when the in-cloud station was inside a cloud). As expected, the remaining interstitial particles exhibited lower CCN activity during a cloud event. The CCN activity can be related to the chemical composition information obtained from aerosol mass spectrometers (an ACSM and an HR-AMS). Positive Matrix Factorisation (PMF) was used to investigate the bulk particle composition for changes related to the air mass origin and potential cloud events. During an observed (south-)eastern European forest fire plume, which lasted for 15 days, the total particle concentration increased from 0.56 µg m ⁻³ to 4.5 µg m ⁻³ . The fire plume particles exhibited a higher CCN activity than those before and after the event?. The forest fire plume event opens the opportunity to compare cloud events occurring in a semi-urban/rural atmosphere typical for the SMEAR IV station with clouds formed under the influence of aged forest fire emissions. |
| Steven | Celik | Georg-August-University Göttingen School of Science | | Studies on the temperature dependent onset of sulfuric acid nucleation | This work is focused on the dynamics of H ₂ SO ₄ particle nucleation in dry supersaturated H ₂ SO ₄ vapor, homogeneously produced by rapid oxidation of SO ₂ through stabilized Criegee-Intermediates from 2-butene ozonolysis. To obtain insight into the mechanisms in early stage nucleation a kinetic model simulates resulting particle size distributions (PSD). By predicting the correct form and modes of the experimentally observed PSD the kinetic model can reliably validate quantum chemically predicted stabilities of small H ₂ SO ₄ clusters since the PSD modes show high sensitivity to the evaporation rates of these smallest clusters (dimer-pentamer). Further enhancement of the temperature module in the simulation now accounts for variation of the reactants temperature during the experiment. This increases the precision of our model to precisely interpret the dynamics of the smallest clusters which are believed to initiate a significant fraction of atmospheric new particle formation events. Additionally the experimental setup has been improved by means of steady control of our batch type reactors temperature within a broader range of operation. The possibility of increasing the reactors temperature paves the way for experimentally determining the onset of particle formation in the gas phase under variable circumstances like ambient pressure and H ₂ SO ₄ concentration. In this work we conduct an experimental series maintaining the properties H ₂ SO ₄ concentration and ambient pressure at magnitudes probed in previous work. The setup allows real time monitoring of ultrafine particles (in a range of 1 nm to 350 nm). For our analysis specific particle sizes were monitored time dependently and are compared to the results of the kinetic model as well as the experimentally obtained final PSD at different temperatures. Our results show a significant temperature sensitivity of the particle formation and are illustratable by particle size traces over time, summarized particle masses and PSD, all plotted against temperature. The depictions can even show temperatures at which the particle formation is inhibited. The results of the simulated PSD are kept in good agreement with the experimentally derived data. |

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| Robin | de Jonge | Lund University | Robin Wollesen de Jonge, Pontus Roldin | Interactive formation and growth of secondary aerosol particles in the marine and boreal forest environment - does marine precursors initiate continental new particle formation? | <p>Biogenic volatile organic compounds (BVOCs) and their oxidation products are known to facilitate the formation and growth of secondary aerosol particles in the ambient atmosphere. Some originate from the ocean, emitted by plankton or bacteria, while others stem from vegetation over the continents. Both marine and continental processes leading to aerosol formation or growth has been researched extensively and described in numerous publications. Their interactions however are yet to be examined. We seek to understand how marine precursors such as dimethyl sulfide (DMS) affects new particle formation over the boreal forest, and how highly oxygenated organic molecules (HOM) originating from monoterpenes grows said particles into the cloud condensation nuclei (CCN) size range.</p> <p>We have utilised the Lagrangian chemistry transport model ADCHEM in reproducing observations from the SMEARII station (61°51' N, 24°17' E) located in Hyytiälä, Finland, during the year of 2018. The model operates along trajectories generated by HYSPLIT, using metrology data from GDAS and incorporating emission inputs from CAMS. The atmospheric cluster dynamics code (ACDC) is coupled to the model in order to consider ion mediated new particle formation from sulphuric acid and ammonia.</p> <p>We demonstrate how ADCHEM captures the gas-phase concentrations of key species including sulfuric acid (SA), HOM monomers and HOM dimers along with new particle formation and growth as observed by CI-API-TOF and SMPS instrumentation, respectively. By running the model without anthropogenic influence we show how DMS-derived SA and ammonia emitted from the ocean is transported inland in quantities sufficient to initiate NPF at the SMEARII station. The newly formed particles grow by condensation of HOM to reach the CCN size range. Our model results also indicate gas-phase concentrations of iodic acid (IA) at approximately 10^{15} molecules cm^{-3}, originating from marine emissions of methyl iodide (MeI). While ADCHEM does not consider the effect of IA on NPF, studies have claimed that IA may be able to cluster is the presence of SA and ammonia. This could increase the marine impact on continental NPF and will become the focus of future work with ADCHEM.</p> <p>We theorise that the interactive marine and continental aerosol formation may act as a key element in the hydrological cycle. Marine air masses not only transport water vapor inland from the ocean but also SA and ammonia that under the influence of HOM formed over the boreal forest initiates the formation and growth of aerosol particles (independent from anthropogenic influence) ultimately leading to cloud formation and precipitation. Consequently, water from the ocean falls as rain where the forest provides BVOCs, HOM and thus the aerosol particles necessary to capture it.</p> |
| Martin Bødker | Enghoff | Danish Technical University, DTU Space | Nir J Shaviv, Henrik Svensmark | Sulphuric acid aerosols in low oxygen environments | <p>On early Archean Earth temperature was high enough for liquid water to exist despite a much weaker Sun - this is known as the Faint Young Sun paradox. One contributing reason may be reduced nucleation due to e.g. lower gas concentrations of either nucleating species or precursors and possibly a lower contribution of ion-induced nucleation due to increased solar magnetic shielding of galactic cosmic rays.</p> <p>To understand this we have to know which nucleation pathways were available in the Archean atmosphere. Here we present the results of experiments on sulphuric acid nucleation in low oxygen atmospheres. Nucleation initiated by photolysis of sulphur dioxide and subsequent reaction between atomic oxygen and sulphur dioxide was measured with a PSM and a separate CPC. The parameters were <10 ppm molecular oxygen with varying levels of sulphur dioxide (4 levels from 40 to 105 ppb), relative humidity (3 levels from 0 to 51%), UV light (254 nm, 4 levels from 55 to 100% power), and ionization (2 levels: Background (~3 per cm^3 per second) and increased w. gamma sources (~42 per cm^3 per second)). We find that nucleation is possible under these conditions and that the measured formation rates correlate positively with all varied parameters. This suggests that the sulphuric acid nucleation system could have played a role in the Archean atmosphere.</p> |

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| Joonas | Enroth | Airmodus Oy | Joonas Vanhanen, Tuukka Petäjä | Development and performance evaluation of a new compact condensation particle counter | <p>A new compact, lightweight, condensation particle counter (A30) was developed, with the aim providing a tool suitable for long term monitoring of indoor and outdoor aerosol particle number concentration, as well as for aerosol research. The new CPC has a small, 0.21 lpm undiluted sample flow rate, and a total of 1.5 lpm inlet flowrate. The excess flow only serves to reduce sampling line losses leading up to the instrument. The small sample flow rate enables the A30 to have a low butanol consumption in the normal operating mode. The measured consumption was found to be 10.4 ml/day, allowing for more than three months of continuous operation using a standard 1 l butanol fill bottle.</p> <p>In laboratory measurements, the A30 was found to have an excellent dynamic range. The particle counter was able to reach high concentrations, up to $1e5 \# \text{ cm}^{-3}$, in single particle counting mode, making it comparable with other commercial high concentration CPCs. In photometric mode, the instrument was found to reach $3e5 \# \text{ cm}^{-3}$ concentrations with more than 90 % agreement with a reference electrometer. The high concentration limit also supports its use in measurements in the urban environment. The instrument is also equipped with both condensate removal functionality, and a user changeable saturator wick, to maintain sustained operation in polluted or humid environments.</p> <p>The new CPC was field tested in a four-week urban measurement campaign at the SMEAR III site in Kumpula, Helsinki (Järvi, et al. 2009). The campaign started on the 18th of January and is scheduled to end on the 18th of February 2022. While writing the abstract, the campaign is still on-going. Based on the initial assessment, the performance of the instrument showed excellent agreement when compared against the stations long term particle number concentration measurement system (TSI 3750). The A30 showed good agreement even during periods of low particle number concentration, due to the non-diluted sampling of the instrument. The A30 is designed to fulfill the requirements of both the CEN/TS 16976 for ambient air total particle number concentration measurements and the upcoming PMP 10nm for vehicle engine emission type approval measurements.</p> |
| Pak Lun | Fung | University of Helsinki | Martha Zaidan, Tuukka Petäjä, Tareq Hussein | Understanding the feature importance in air quality black-box models | <p>Air pollution has been a global challenge to public health, especially in urban cities where more than 90% of the population lives and suffers from poor air quality. In the past decades, researchers have been looking for solutions by understanding the interaction and the sources of the various pollutants. Apart from physics-based models that require costly computational resources, the use of data-driven models have been on the rise. Among all data-driven models, black-box models, including neural networks and random forest, manage to give high accuracy but low transparency of their internal workings. It can be difficult for both the model creators and the model users to understand why some particular outcomes were reached and what leads to such outcomes.</p> <p>In this abstract, we aim to find a way to explain and understand the feature importance in air quality black-box models by comparing some model-agnostic interpretation methods. These models include the inclusion of local surrogate models (LIME), the calculation of shapley values, the use of counterfactual explanations, etc. Both LIME and shapley values are attribution methods; the former explains a prediction by replacing the complex model with a locally interpretable surrogate model while the latter estimates a feature value by replacing it with a random value. On the other hand, counterfactual explanations is an example-based method, which describes the smallest change to the feature values that changes the prediction to a predefined output.</p> <p>Extending from a study on the evaluation of black carbon white-box and black-box models, we use the black carbon dataset as a case study to compare and demonstrate which model-agnostic interpretation methods work the best for each data-driven models. This study can then serve as a benchmark for similar air quality models. This also helps exploring new air quality models with enhanced accountability and shows great potentials to develop these models as virtual sensors.</p> |
| Humaira | Ghayas | CSIR-National Physical Laboratory; Academy of Scientific and Innovative Research (AcSIR) | Humaira Ghayas, Sachchidanand Singh | Global photosynthetically active radiation and its relationship with global solar radiation in Delhi | <p>Photosynthetically Active Radiation (PAR) and broadband solar radiation (SW) are the enigmatic and indispensable parameter in studies including climate change, plant canopy radiation regime, remote sensing of vegetation, plant physiology, photosynthetic process, carbon fixation and biomass production. Despite of the important role played by the PAR flux worldwide measurement networks are lacking. In the present study, 18-years data (January 2001 to December 2018) of hourly PAR and SW radiation were downloaded from Clouds and the Earth's Radiant Energy System (CERES) for Delhi city. The data were used to find the temporal variability of PAR along with its dependency on sky conditions. Annual mean value of 0.44 obtained for daily values of PAR/SW ratio which was favourably compared with past PAR/SW ratio values reported in the literature. Seasonal variation of PAR/SW ratio ranges from 0.472 ± 0.016 during monsoon to 0.424 ± 0.017 during winter, attributable to change in local air mass climatology. PAR/SW ratio ranges from 0.22 to 0.65 on hourly basis and 0.38 to 0.55 on daily basis as the sky condition changes from clear to overcast. Analysis of hourly values revealed the daily pattern with lower value around noon whereas higher values in morning and afternoon hours. Finally, the effect of aerosol loading incurs substantial changes in PAR/SW ratio.</p> |

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| Paul | Glantz | Department of Environmental science, Stockholm University | Olesugun Gabriel Fawole, Johan Ström, Martin Wild, Kevin Noone | Unmasking the effects from aerosols on warming over Europe | <p>MERRA-2 reanalysis aerosol optical thickness has decreased substantially in Europe in the summer half year (April – September) since 1980, up to near 50% in Central and Eastern Europe. ERA5 reanalysis cloud cover from European Centre for Medium-range Weather Forecasts has as well decreased during the period 1979 – 2020. At the same time, strong positive trends in ERA5 surface solar radiation downwards for all-sky and clear-sky conditions (SSRD and SSRDc, respectively) and temperature at 2 m (T2m) are found for Europe in summer. ERA5 T2m agree well with surface temperature measured at 43 in-situ sites in Europe. Global Energy Balance Archive ground-based measurements and the Santa Barbara Disort Atmospheric Radiative Transfer model show also strong increases in SSRD and SSRDc, respectively, during the latest four decades. Estimates of warming caused by the increases in incoming solar radiation are based on energy budget approximations and the Stefan Boltzmann law. The increases in near surface temperatures, due to the decline in aerosols, are substantial for clear-sky conditions: up to about one degree for Central and Eastern Europe. Warming estimated for all-sky conditions is somewhat higher. This means that other regions on earth with high anthropogenic aerosol loadings today will probably experience significant effects on the warming if aerosol concentrations are to decrease in the future. Furthermore, the total warming for clear-sky conditions is nearly double the global mean temperature record of about 1.1 °C over large parts of Europe, while somewhat lower for all-sky conditions. The effects from aerosols on warming over the southerly Iberian Peninsula is weaker compared to countries further north. The fast warming over Iberian Peninsula is probably caused by greenhouse warming, tropical expansion, more arid surfaces, reduced cloud cover and decline in aerosols. We show in this study that decline in anthropogenic aerosols can certainly not explain all warming observed and particularly not considering Iberian Peninsula. Anthropogenic aerosols over large part of Europe have then temporarily masked fast warming from increases in greenhouse gases. CO2 from fossil fuels is of particularly serious concern, since it can continue to affect climate for thousand years. The large regional effect found in this study suggests also the importance for improvements of descriptions of climate drivers in regional climate models to realistic assessing future climate change.</p> |
| Roope | Halonen | Center for Joint Quantum Studies and Department of Physics, Tianjin University | Roope Halonen | Multicomponent clusters: Building a bridge between quantum chemistry and classical nucleation theory | <p>Nature is quantum mechanical. Indeed, in the field of atmospheric aerosol research, quantum chemical calculations have proven to describe the thermodynamics of sub-nanometer clusters more accurately than the liquid drop model used in classical nucleation theory (CNT). However, the standard quantum chemical free energies of multicomponent clusters are fundamentally incompatible with the quantity appearing in the exponential of the CNT nucleation rate expression. The origin of this incompatibility is known to be in the statistical thermochemistry, but it is also connected to a much debated issue of self-consistency within CNT. Although these issues do not affect the main results of previous kinetic studies of nucleation using quantum chemical data (e.g., ACDC simulations), a thorough analysis and discussion about the definition of free energy in quantum chemistry of cluster formation has been missing.</p> <p>By presenting a consistent definition for the formation free energy (using the basic principles from statistical mechanics and nucleation theory), we are able to integrate the high-level thermochemical data to the basic framework of CNT and the liquid drop model contained therein. Unlike some earlier “semi-CNT” models where a quantum correction is applied on the liquid drop thermodynamics, the presented approach yields a proper “first principle” nucleation theory for multicomponent systems. Due to the assumptions inherited from CNT, the obtained nucleation rates correspond to steady state and conditions where cluster growth is mainly monomeric. The analytical results given by the presented theory are fully compatible with the state-of-the-art numerical ACDC simulations carried out for an identical system.</p> <p>Detailed cluster growth simulations, such as ACDC, are fundamentally superior, but using them can be technically complicated and require methodological and computational expertise. The suggested theory provides the means for quick “back-of-the-envelope” estimates for the nucleation rates. Also, extending the cluster space to ternary clusters (and beyond) might result in numerically stiff, or unstable, set of equations inside the ACDC model. Whereas the analytical expression for the steady-state nucleation rates provide a general solution for arbitrarily complex systems.</p> |

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| Eemeli | Holopainen | Finnish Meteorological Institute | Eemeli Holopainen, Harri Kokkola, Celia Faiola, Anton Laakso, Thomas Kühn | Plant stress emissions reduce aerosol radiative forcing | <p>The majority of the emitted biogenic volatile organic compounds (VOCs) are produced by terrestrial vegetation, such as trees and plants. Biotic plant stress, such as herbivore infestation, is predicted to increase VOC emissions of trees. This increase can lead to enhanced formation of biogenic secondary organic aerosol (SOA) particles, which further can influence cloud formation and radiative effects. This is due to direct and indirect climate effects of aerosol particles. The contribution of biotic stress to SOA formation is not usually considered in global aerosol-climate models and thus more investigation is needed here to properly evaluate the effects of SOA to climate. In this study, we used the ECHAM-HAMMOZ (ECHAM6.3-HAM2.3-MOZ1.0) global aerosol-climate model, with sectional aerosol characterization SALSA, to simulate the effects of biotic stress, due to herbivore infestation, on needleleaf evergreen boreal and broadleaf deciduous boreal trees, and the subsequent effects on SOA formation, clouds and radiative effects. This was done by altering the fraction of stressed plants and analysing their effect on SOA burden, cloud droplet number concentration at cloud top and all-sky and clear-sky shortwave radiative forcing (RF). In the global model simulations we used MEGAN v2.1 emission model to simulate how the monoterpene emissions from boreal trees change with increasing herbivore infestation level. We found out that increasing the extent of stress in needleleaf evergreen boreal and broadleaf deciduous boreal trees increased the SOA burden by 120 % at maximum. This indicates that increasing the VOC emissions due to biotic stress enhances the SOA formation. In addition, CDNC at cloud top increased, with increasing biotic stress, by maximum of 40 %. This indicates that increasing SOA formation increases the amount of particles acting as cloud condensing nuclei. Increasing the biotic stress also decreased both all-sky and clear-sky RF. The strongest RF was achieved with the simulation where all of the plants were stressed and for clear-sky case the RF was approximately -4.3 W/m² and for the all-sky case approximately -3 W/m². This was due to increase in SOA particle concentrations at a certain size range that enhances the scattering of incoming solar radiation back to space. These results indicate that including the effect of biotic stress on plants can have a considerable effect on SOA formation, clouds and radiative effects and could be one of the means to reduce uncertainties in estimating the radiation budget of the Earth.</p> |
| Noora | Hyttinen | University of Jyväskylä | Noora Hyttinen, Iida Pullinen, Aki Nissinen, Siegfried Schobesberger, Annele Virtanen, Taina Yli-Juuti | Comparison of computational and experimental saturation vapor pressures of α -pinene + O ₃ oxidation products | <p>Accurate information on gas-to-particle partitioning is needed to model the formation of secondary organic aerosol (SOA). However, measuring saturation vapor pressures (which are often used to derive gas-to-particle partitioning coefficients) of atmospherically relevant multifunctional organic compounds is extremely difficult.</p> <p>Many studies have shown that oxidation products of α-pinene are able to form SOA in the presence of seed particles. We estimated saturation vapor pressures of α-pinene ozonolysis-derived SOA constituents using the Filter Inlet for Gases and AEROSols coupled with a chemical ionization mass spectrometer (FIGAERO-CIMS) and CONductor-like Screening MOdel for Real Solvents (COSMO-RS). Using FIGAERO-CIMS, saturation vapor pressures are estimated from measured desorption temperatures of compounds present in SOA samples collected on filters. For our COSMO-RS calculations, we selected 1-7 different potential isomers for each elemental composition, since a mass spectrometer is not able to provide detailed information about the isomers that are detected.</p> <p>We found a good agreement between experimental and computational saturation vapor pressures for molecules with molar masses around 190 g/mol and higher, most within a factor of 3 comparing the average of the experimental vapor pressures and the COSMO-RS estimate of the isomer closest to the experiments. Smaller molecules likely are too volatile to be measured using our experimental setup, either because the molecules are not condensing on the aerosol particles or they evaporate from the sample before the thermal desorption experiment begins. On the other hand, FIGAERO-CIMS cannot be used to estimate saturation vapor pressures of extremely/ultra low volatility compounds because those compounds do not evaporate from the condensed-phase sample even with strong heating. The lowest experimentally determined saturation vapor pressure among the molecules selected for our study was $8 \cdot 10^{-11}$ Pa.</p> <p>The molecules with molar masses below 190 g/mol that have several orders of magnitude difference between the computational and experimental saturation vapor pressures observed in our experiments are likely products of thermal decomposition occurring during thermal desorption. For example, dehydration and decarboxylation reactions are able to explain some of the discrepancies between measured and calculated saturation vapor pressures. Based on our estimates, FIGAERO-CIMS can best be used to determine saturation vapor pressures of compounds with low and extremely low volatilities, while COSMO-RS can be used to estimate saturation vapor pressures of any molecule with up to tens of non-hydrogen atoms.</p> |

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| Ulas | Im | Aarhus University, Department of Environmental Science | Susanne E. Bauer, Lise M. Frohn, Camilla Geels, Kostas Tsigaridis, Jorgen Brandt | Present and future PM2.5-related premature mortality | <p>The Economic Valuation of Air Pollution (EVA) system has recently been used estimate the present (2015) and future (2015-2050) global and regional PM2.5-related premature mortality. Surface concentrations are simulated using the fully-coupled Earth system model, GISS-E2.1, using future emission projections SSP1-2.6, SSP2-4.5 and SSP3-7.0. In addition, we have tested the sensitivity of PM2.5-related mortality estimates to model bias, emission reductions and ageing population.</p> <p>The all-cause PM2.5-related global premature mortality is estimated to be 4.4 million, while the cause specific GEMM model estimated a global PM2.5-related mortality of 4.3 million. Ischemic diseases are found to be the leading cause of PM2.5-related premature deaths, contributing by 23% to 62% of deaths regionally, and by 35% globally. The global burden of premature deaths is mainly driven by the emission hotspots in Asia, which in 2015 alone contribute with 3-3.3 million. Bias correction for the simulated PM2.5 concentrations in 2015 led to an increase of up to 41% to 73% in the global PM2.5-related premature mortality, in the non-linear and linear models, respectively. Finally, while results show that PM2.5-related premature mortality decreases by up to 57% due to emission reductions by 2050 alone. Taking the projected increase and ageing of the population into account leads to increases of premature mortality by up to a factor of 2. This implies that the population and its age composition exposed to the surface PM2.5 can be more important than the level of PM2.5, which is an important challenge for the future.</p> |
| Jonas | Jakobsson | Lund University | Erik Ahlberg, Pontus Roldin, Robin Wollesen de Jonge, Adam Kristensson, Erik Swietlicki | Exploring machine learning as a tool to investigate the impact on Swedish air quality during the covid-19 global shutdown | <p>It would be fair to assume that the 2020 covid shutdown is the largest un-planned environmental intervention experiment to date. All over the world people have been forced to dramatically change their way of life. The worldwide restrictions have altered consumer patterns, shut down businesses and industries, limited traveling both locally and globally and changed our energy production and needs. This could be expected to lead to a fast and significant change in global aerosol emission patterns. However, there are several issues that complicate the interpretation of the impact on air quality, especially based on collected air quality data.</p> <p>To investigate the impact of the shutdown on Swedish air quality we have adapted a machine learning approach. The aim has been to construct a counterfactual "business as usual" scenario, to which we can compare observations of air quality data collected at the ACTRIS/ICOS site Hyltemossa in southern Sweden during the shutdown. This approach has previously been applied by other groups, but in more densely populated areas with vastly different levels of ambient air pollution compared to typical Scandinavian conditions. A random forest ensemble learning method was used to train models to predict air quality parameters from meteorological data. The training parameters were meteorological parameters collected locally at the Hyltemossa field station (air temperature, relative humidity, wind speed, wind direction) and origin of air mass data derived from HYSPLIT-back trajectories, expressed as the time over different geographical regions during the last 24 h, 48 h, 72 h, 1 week and 2 weeks, including cumulative precipitation and solar radiation upwind Hyltemossa based on these trajectories. The models were trained to predict several different air quality variables, including PM1, PM2.5, PM10, BC, ACSM and other air quality parameters. A training dataset was compiled for the period from 2018-01-01 to 2021-01-01, to the extent air quality data was available. The data for the strictest covid shutdown period was excluded from the training dataset and predicted with the models to construct a counterfactual "business as usual" scenario. The counterfactual scenarios were then compared to the observed air quality data to investigate the presence of a "covid effect". Preliminary results indicate that the method may detect deviations between the observed air quality data during the period and the prediction for a "business as usual" scenario, although the effect does not appear to be dramatic. Further development of the models, validation and analysis of the initial findings are ongoing to evaluate the direct effect on Swedish air quality of the shutdown, and in extension also more complex effects on atmospheric chemistry and secondary pollutants. We gratefully acknowledge ACTRIS Sweden and ICOS Sweden for extensive work in producing and sharing meteorological and air quality data used in this project.</p> |

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| Adam | Kristensson | Lund University | Adam Kristensson, Adrian Reyes Martin, Hugo Savill Russell, Robin Wollesen De Jonge, Sadath Ismayil, Pontus Roldin, Mikko Sipilä, Andreas Massling, Matthew S. Johnson. | The aerosol chase | <p>Determining how aerosol particle number size distributions develop during long range transport, due to atmospheric chemistry and aerosol processes, and how aerosol sources contribute to this aerosol population is one of the greatest mysteries in aerosol science. One way to approach this issue is to follow the movement of the aerosol with the wind, and measure the aerosol particles and the precursor gases as they develop in this air mass, which is called the “Lagrangian” approach.</p> <p>In this trial Lagrangian project, measurements are made using a mobile platform with a road vehicle. Aerosol particle number size distributions are measured with an SMPS system, together with nitrogen oxides, and carbon dioxide. The instruments are powered with a battery-inverter system, which allows operation with the engine off, to avoid contamination. To avoid non-isokinetic sampling errors the measurements are performed when the vehicle is stationary at short stops. The measurements are performed a few kilometers away from major emission sources, to focus on regional effects and avoid short-time variability. However, the Lagrangian path still passes through major emission sources such as towns and highways.</p> <p>Two successful test runs have been performed during spring 2021 in the first set of measurements. The chases were initiated from southernmost Sweden, where the air was followed for several hours until the battery to support power to the instruments was depleted. Measurements in the car were validated with the ACTRIS infrastructure station Hyltemossa data. After analysis of this first set of experiments, the next stage of measurements will be performed during 2022, possibly also including a Lagrangian path passing through Denmark and Sweden at the same time.</p> <p>Modelling with a Lagrangian aerosol dynamics and chemistry model is necessary to study the effects of coagulation, condensation, chemistry air mixing, and to quantify the source contributions from various new emissions along the trajectory path. Qualitatively, however, it is anticipated that these processes can be understood and identified with the naked eye by observing changes in the measured parameters along the wind path. If the results of this pilot project are successful, they will mean a revolution in aerosol science in terms of understanding the in situ dynamics of aerosol populations. Once validated, the methodology can then be used worldwide.</p> |
| Jakub | Kubecka | Aarhus University | Jakub Kubecka, Anders S. Christensen, Freja R. Rasmussen, and Jonas Elm | Atmospheric cluster formation: speed-up by employing quantum-machine-learning methods | <p>Atmospheric aerosol particles have a crucial cooling effect on the Earth’s temperature. However, their contribution also introduces the largest uncertainty in the current global climate modeling. This uncertainty is caused by the lack of understanding on how aerosol particles are formed and grow to larger sizes. Therefore, we focus on studying molecular-cluster/new-particle formation in the atmosphere. This involves the study of the collision of single molecules in the atmosphere that forms molecular clusters stable enough to grow by condensation of other molecules into larger particles, aerosols.</p> <p>The most tedious part of this study is the evaluation of the cluster stabilities. With the enormous number of cluster configurations and possible combinations of molecules, this task becomes extremely computationally demanding. One must perform configurational sampling for each molecular cluster size and find the lowest free energy structures that would most likely be present in the atmosphere. Accounting for all possible fragmentations, we use balance equation to calculate the cluster evaporation rates. These rates are thus exponentially dependent on the free energy changes during the evaporation, and therefore, accurate quantum chemistry (QC) methods are required. The higher accuracy of the QC method, the greater the computational cost.</p> <p>We present a quantum-machine-learning (QML) technique that can be utilized to produce an accurate prediction of cluster binding energies. We show that trained QML can achieve errors lower than 0.5 kcal/mol on the sulfuric acid-water cluster systems. Additionally, we show prediction of binding energies for larger clusters that were not included in the training set with only a slight loss in the prediction accuracy. However, this is an enormous gain in the computational efficiency as it can accelerate both the configurational sampling and the cluster stability evaluation. These are very encouraging results for further studies of other atmospheric acid-base molecular clusters.</p> |

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| Xinyang | Li | Institute for Atmospheric and Earth System Research (INAR), University of Helsinki, Helsinki, 00560, Finland | Xinyang Li, Lubna Dada, Chao Yan, Nina Sarnela, Pauli Paasonen, Risto Makkonen, Markku Kulmala, Tuomo Nieminen | PARAMETERIZATION OF ATMOSPHERIC ULTRAFINE AEROSOL FORMATION RATES IN TWO CONTRASTING ENVIRONMENTS: A BOREAL FOREST AND A MEGACITY | <p>Atmospheric new particle formation (NPF) events have been proven to have great impact on human health and climate. The parameterizations for the formation rate on particles in smaller size ranges (i.e. J1.5, J2, J3, etc.) were developed extensively. However, the parameterizations focusing on larger particle sizes lacks implementation and simplicity for global models. Here, we present the parameterization for particle formation rates between 5-9 nm in two contrasting environments. Including such contrasting environments into the parameterization will help expand the applicability of the models in various environment in global scales.</p> <p>In this study, we parameterize 5-9 nm particle formation rates (J) for Hyytiälä (SMEAR II station, 2016-2019) and Beijing (Beijing University of Chemical Technology west campus 2018-2020). The parameterizations of formation rates are based on the analysis of atmospheric particle number-size distribution measurements measured using DMPS. The variables involved in the parameterizations are sulphuric acid (SA), temperature (T), relative humidity (RH) and condensation sink (CS). Sulphuric acid (SA) was measured using CI-API-ToF mass spectrometer, CS and J were calculated applying the methods presented in Kulmala (2012).</p> <p>Several functional forms were considered for the parameterizations involving different combinations of the variables listed above. All models were developed applying 5 oC temperature windows from T < -5 oC to T > 20 oC (7 T windows applied), the preliminary parameterization results were listed below: Model (1): $J5 = k0 \times [H2SO4]$ ($r = 0.73$, $p < 0.05$) Model (2): $J5 = k0 \times [H2SO4] \times RH^{k1}$ ($r = 0.8$, $p < 0.05$) Model (3): $J5 = k0 \times [H2SO4] \times RH^{k1} \times CS^{k2}$ ($r = 0.84$, $p < 0.05$)</p> <p>The result showed an underestimation of the parameterized formation rate especially for Beijing when considering only SA into the model. The parameterizations result shows improved correlation coefficient of the model by adding RH and CS into the model with exponent coefficients, while no data underestimation is observed.</p> <p>These results showed good performance of the model involving RH and CS using data from contrasting environments, which would potentially fill the gap in particle formation rate parameterization in larger particle sizes. Thereupon, these results bring possibility of testing data from other environments (i.e. urban area and Mediterranean region) on the models for the preparation to be implemented into global aerosol models.</p> |
| Yuanyuan | Luo | University of Helsinki | Olga Garmash, Haiyan Li, Frans Graeffe, Arnaud P. Praplan, Anssi Liikanen, Yanjun Zhang, Melissa Meder, Otso Peräkylä, Josep Peñuelas, Ana María Yáñez-Serrano, Mikael Ehn | Oxidation product characterization from ozonolysis of the diterpene ent-kaurene | <p>Terpenes are the primary component of biogenic volatile organic compounds (BVOCs) in the atmosphere, accounting for 40 – 65% of the total BVOCs emission. Once emitted into the atmosphere, terpenes can react with different oxidants and form a wide range of organic species. Many of these species have low enough volatilities to condense onto aerosol particles, making terpenes an important precursor for secondary organic aerosol (SOA). To date, there has been a number of studies investigating monoterpenes and sesquiterpenes. However, the potential atmospheric importance of larger terpenes, like diterpenes, remains unclear. Therefore, this study set to explore the characteristics of diterpene ozonolysis under atmospherically relevant conditions. During the experiments, different concentrations of the diterpene ent-kaurene and ozone were injected into the 2 m³ COALA chamber. Seed particles were also added to provide a condensation sink for low-volatile oxidation products. The Vocus proton transfer reaction time-of-flight mass spectrometer (PTR-TOF) and a nitrate-based chemical ionization atmospheric pressure interface time-of-flight mass spectrometer (CI-API-TOF) were utilized to measure the precursor and oxidation products. In this study, we detected various products of kaurene ozonolysis with varying oxidation levels, including highly oxygenated organic molecules (HOM), both monomers and dimers, for the first time. The molar yield of HOM from kaurene ozonolysis was estimated to be a few percent, which is similar to those estimated for many monoterpenes and sesquiterpenes. The most abundant monomers that nitrate CI-API-TOF measured were C19H28O8 and C20H30O5, and dimers were C38H60O6 and C39H62O6. These HOM products are assumed to be LVOCs (low-volatility organic compounds) or ELVOCs (extremely low-volatility organic compounds), and they readily condensed to the particle phase. Fragment distributions of kaurene in the Vocus PTR instrument were also quantified. Our findings highlight the similarities and differences between diterpenes and smaller terpenes during their atmospheric oxidation, but more studies on different diterpenes are needed for a more comprehensive view of their importance in the atmosphere.</p> |

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| Vilhelm | Malmborg | Lund University | VILHELM B. MALMBORG, François-Guillaume Ide, Dan Madsen, Ioannis Sadiktsis, Michaël Toublanc, Patrick van Hees, Andrew Grieshop, Joakim Pagels | Application of a cone calorimeter set-up for controlled emission studies of biomass pyrolysis and combustion | Systematic study of the connection between combustion and properties of particles emitted during diverse biomass combustion types is generally limited by the highly variable combustion conditions within and across burns. Flow reactor experiments with simple fuels (e.g., benzene) can reduce variability but does not directly represent biomass combustion emissions. In this study, we bridged this gap by combustion in a cone calorimeter according to ISO 5660-5 to control total heat flux (HF) and gas (air/N ₂) flow rates to simulate distinct combustion conditions, pyrolysis, well- and under- ventilated combustion. We conducted over 40 experiments with widely ranging HF and flow conditions while monitoring fuel mass loss to quantify emission yields. An Aerosol Mass Spectrometer (AMS), a multi-wavelength aethalometer and a particle size spectrometer (DMS5000) measured time-resolved evolution in particle properties during burns. We also collected filters for offline analysis of OC/EC, PAHs/oxy-PAHs and UV-Vis absorption by methanol extracts. Pyrolysis conditions (in N ₂) generated high primary OA emission factors (0.03-0.5 g/g). Absorption Ångström Exponents (AAE) were between 2-3 for pyrolysis conditions. For high-flow combustion, higher HF increased eBC emissions (0.005 to 0.01 g/g), decreased AAE (1.7 to 1.2), and reduced OA emissions two orders of magnitude compared to pyrolysis conditions. Low-flow and oxygen-starved combustion was associated with extreme PAH emission factors up to 0.1 g/g (~100 times higher than under well-ventilated conditions). Further analysis of the data set will parameterize emissions based on conditions underlying diverse biomass combustion modes: heat release rate, oxygen content, stoichiometry, and initial conditions such as HF, moisture content or air flow rate (cooling). With this we aim to unify earlier observations collected at different scales, for example from cook stoves, wild fires, and heating stoves. |
| Emil | Mark Iversen | Aarhus University | Henrik B. Pedersen, Merete Bilde | A New Method for Assessing Particle Wall Loss in the AURA simulation Chamber | Simulation chambers are widely used to mimic atmospheric processes hereby increasing our knowledge of the atmosphere. One of the experimental challenges of using atmospheric simulation chambers is the additional loss of particles and gases to the walls that enclose the volume of the simulation chamber. Since the atmosphere has no walls, this loss to the chamber walls can pose limitations to laboratory studies if not properly accounted for. The particle wall loss in simulation chambers depends strongly on both particle properties as size, mass, and charge and simulation chamber properties as the surface-to-volume ratio of the simulation chamber, the local electric field, and the explicit flow conditions within the simulation chamber. This may even lead to significant time dependencies of the particle wall loss during observations since both particle and chamber properties typically change as the experiment proceeds. This includes growth or disintegration of the particles, variation of the particle charge distribution induced by cosmic rays, and the change of the chamber volume as the attached instruments sample air from the simulation chamber. With this study, we aim to investigate the possibility to simplify the corrections for particle wall loss and potentially eliminate unobservable time-dependencies of the particle wall loss. The central ideas are to actively measure the volume of the atmospheric simulation chamber during the experiment and to induce a strong mixing of the chamber atmosphere. The mixing is carried out with a set of custom-made stainless steel fans, which are installed on each side of the Aarhus University Research on Aerosols (AURA) atmospheric simulation chamber. A set of two laser distance sensors are used to measure how the AURA chamber shrinks when air is sampled by attached instruments. These laser distance measurements are calibrated to yield the volume of the AURA chamber, resulting in time-resolved volume data of the AURA atmospheric simulation chamber. |

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| Andreas | Massling | Aarhus University | Jakob Boyd Pernov, Peter Tunved, Sangeeta Sharma, Eija Asmi, Niku Kivekäs, Johan Ström, Julia Schmale, Hans Christian Hansson, Henrik Skov | Trends of particle number in segregated size fractions at Arctic continental and high Arctic sites | <p>Arctic temperatures are increasing two to three times as fast as in the rest of the world, making the region most severely affected by climate change; a phenomenon known as Arctic amplification. Aerosols are key players here, influencing the radiative balance by both, directly (scattering and absorption) and indirectly (serving as cloud condensation nuclei; “CCN”). In a changing Arctic climate, the presence and transport pathways of aerosols will be affected, ultimately impacting the radiation balance of the atmosphere and creating feedback mechanisms for aerosol formation and emissions of precursor gases. Trends of atmospheric particle number concentrations in segregated size fractions are an indicator of the potential changes linked to direct and indirect aerosol effects.</p> <p>Particle number size distributions (originating from Scanning and Differential Mobility Particle Sizers; SMPS/DMPS) were used from the different Arctic continental and high Arctic sites: Pallas (northern Finland), Tiksi (Siberian Arctic), Villum Research Station (northeastern Greenland), Alert (Canadian Archipelago), and Zeppelin Observatory (Svalbard). The study comprises data between 2010 and 2019 depending on the measurement site.</p> <p>Particle number concentrations in segregated size fractions representing nucleation (10-35 nm), Aitken (35-80 nm) and accumulation (80-300 nm) mode aerosols were calculated by integration of the size distributions. The trends in number concentrations were investigated using the Mann-Kendal test and Theil Sen slope estimator on the 90th % confidence interval via the 3 pre-whitening algorithm. Data were aggregated into daily medians and segmented into the meteorological seasons as these typically identify different origins and formation and transformation processes.</p> <p>As a major result, it was found that all high Arctic stations (Villum, Alert and Zeppelin) show varying trends in direction and magnitude (between 2010 and 2019) for the different segregated size fractions that were investigated. Similarly, such results were found for the Arctic continental sites Tiksi and Pallas. The stations (Arctic continental as well as high Arctic) are located in different climatic regimes, which might be the main reason for this finding. The study underlines the complexity of source regions, atmospheric processing and probably meteorological changes that contribute to the observation of specific particle number size distributions at measurement sites located within the Arctic.</p> <p>This work significantly contributes to a more thorough understanding and interpretation of particle number size distributions and their possible changes in Arctic continental and high Arctic locations and offers insights into climatic implications of particle physical properties within a future Arctic atmosphere.</p> |
| Tinja | Olenius | Swedish Meteorological and Hydrological Institute | Daniel Yazgi, Tinja Olenius | A flexible tool to incorporate aerosol formation rate predictions from molecular models in large-scale models | <p>Formation of secondary aerosol particles from condensable vapors is a common atmospheric process that has significant but uncertain effects on aerosol particle number concentrations and aerosol-cloud-climate interactions. Assessing new-particle formation rates for different chemical compounds is an active field of research, with new data constantly becoming available from experimental and molecular modeling studies. In general, models benchmarked against measurements provide a straight-forward means to assess formation rates over a wide range of atmospheric conditions for the given species. The formation rate is a function of several parameters, including the concentrations of the contributing vapors, the temperature, and the molecular cluster scavenging sink due to larger aerosols. Depending on the chemical compounds, the formation rate may also depend on atmospheric ion production rate or relative humidity. The formation rate data can be implemented in large-scale climate and air quality models as parameterizations or look-up tables. While deriving reliable, optimal parameterizations becomes cumbersome with an increasing number of independent parameters, the benefit of look-up tables is that no pre-processing of data is needed, and values determined from a table of sufficiently high resolution are always close to the original data. In this work, we build a tool to incorporate molecular modeling data in atmospheric models by flexible routines to generate and interpolate look-up tables of new-particle formation rates. The table generator applies by default a molecular cluster dynamics model with quantum chemistry input, and the interpolation routine uses a multivariable interpolation algorithm, applicable to different numbers of independent parameters. Formation rates from parallel formation pathways due to separate chemical mechanisms are summed when there exists more than one table. This approach covers the following aims: (1) flexible implementation of molecular modeling results in large-scale models, (2) inclusion of arbitrary chemical compounds for which there exist chemical data, (3) automatic multivariable interpolation, and (4) user-friendly routines with no need for modifications depending on, for example, included chemical compounds. These routines facilitate the implementation and testing of different aerosol formation rate predictions in large-scale models, enabling straight-forward inclusion of new or updated data sets, without the need to apply separate routines for different types of data that involve different chemistries or other parameters.</p> |

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| Daria | Pashneva | Center for Physical Sciences and Technology, Saulėtekio av. 3, LT-10257 Vilnius, Lithuania | Inga Garbariene, Julija Pauraitė, Agnė Minderytė, Vadimas Dudoitis, Lina Davulienė, Kristina Plauškaitė-Šukienė, Vidmantas Ulevičius, Vitalij Kovalevskij, Mindaugas Gaspariūnas, and Steigvilė Byčenkienė | Impact of ambient submicron particulate pollution on air quality in mechanically ventilated office buildings | Submicron particulate matter (PM1) are of great importance due to their significant impacts on air quality, human health, and climate change. PM1 generated from various anthropogenic (biomass burning, fossil fuel combustion) and biogenic sources can be transported to indoor space, where people spend a lot of their time (85-90 %). Urban PM1 commonly consists of inorganic ions (sulfate, nitrate, and ammonium), carbonaceous species (organic compounds and black carbon), and trace metals. In low-energy airtight buildings, PM1 is transferred to the indoor environment mainly through the ventilationsystem; therefore, the penetration of PM1 depends on the system filtration efficiency. To evaluate the impact of outdoor PM1 pollution on indoor air quality, the chemical composition and sources of submicron particulate matter were simultaneously investigated in a mechanically ventilated office building in Vilnius, Lithuania. The measurement campaign was carried out during the heating period from October 15, 2020 to February 8, 2021. The investigated space was equipped with a three-stage air filtration system (G4-F7-F9). Near real-time measurement of indoor/outdoor chemical composition was performed by an Aerosol chemical speciation monitor (ACSM (organic matter, sulfate and nitrate)) and Aethalometer (eBC). In parallel with online measurements, the filter-based elemental composition of PM1 was investigated by broad-beam particle-induced X-ray emission (PIXE). In this work, the enrichment factor (EF) was adapted to distinguish whether the trace element (TE) in indoor/outdoor PM1 was derived from natural and anthropogenic origin. Organic matter was the dominant fraction of PM1 with an average concentration of 7.31 µg/m3. The mean mass concentration of sulfate, nitrate, and eBC were 3.74 µg/m3, 2.5 µg/m3 and 1.93 µg/m3, respectively. The sum of total trace elements comprised the smallest contribution to outdoor PM1, with an average mass concentration of 0.16 µg/m3. The results of the complex source apportionment demonstrated a dominant contribution of fossil fuel combustion to eBC, while the organic compounds were mostly from biomass burning emissions. According to the EFs, the elements Al, Si, Ca and Fe were considered to be of crustal origin, while Sc, Co, Ni, Cu and Zn were recognized as anthropogenic in origin. K and Mn were associated with mixed sources. Comprehensive chemical analysis of PM1, including carbonaceous matter and trace elements, allowed us to quantify the contribution of various sources of particulate pollution and their impact on personal exposure. The concentrations of chemical species of PM1 in indoor air were significantly lower than those in outdoor air. The median I/O ratios (which represented the penetration of PM1) of all investigated PM1 components were low (I/O < 0.12). These results indicate that the three-stage building filtration system (G4-F7-F9) has high protection against particulate pollution of different origin and could markedly reduce exposure to PM1. |
| Daria | Pashneva | Center for Physical Sciences and Technology | Steigvilė Byčenkienė, Ieva Uogintė, Julija Pauraitė, Agnė Minderytė, Lina Davulienė, Kristina Plauškaitė, Martynas Skapas, Vadimas Dudoitis, Jelena Andriejauskienė, Vidmantas Ulevičius, Dalia Jasinevičienė, Valda Araminienė, Valda Gudynaitė-Franckevičienė, Iveta Varnagirytė-Kabašinskienė, Eugenija Farida Dzenajavičienė, Nerijus Pedišius, Egidijus Lemanas, Tomas Vonžodas, Pierre Sicard | The anthropogenic black carbon emissions and its deposition on leaves and needles in the Baltic region | It has been estimated that anthropogenic BC emissions from transport and residential combustion are 24% and 60%, respectively and it is the main sources of BC in urban areas. In recent years, BC emissions from residential combustion would increase due to insufficient regulation and use of old boilers. The purpose of the study is to consider the potential impact of urban trees on the removal of BC by common spruce and birch, as well as on the formation of BC, mass concentration and distribution of sources. Measurements were performed during July-October 2020 at an urban background station in Vilnius. BC aerosol particles were found in all samples of leaves and needles as a result of the deposition. Larger agglomerates were found on the needles of conifers than on broadleaf trees, as they form a thicker epicuticular wax layer and are more efficient in accumulating BC. The diameter of primary particles ranged from 32.45 nm to 44.13 nm on spruce needles. Tree species in urban environment can help improve air quality by removing BC from the air. The studies on the combustion of solid biomass fuels in modern and old heating systems showed that BC emissions are significantly higher in old systems. This work was carried out in the framework of the project "Investigation of aerosol black carbon emissions from biomass incineration units and deposition on tree foliage" (RTO Lithuania). |
| Julija | Pauraitė | State research institute Center for Physical Sciences and Technology | Julija Pauraitė, Agnė Minderytė, Vadimas Dudoitis, Kristina Plauškaitė and Steigvilė Byčenkienė | Influence of urban submicron particles on atmospheric radiative balance during high pollution event | Complex observational studies of aerosol properties under real and various conditions are crucial for reducing uncertainties in estimation of aerosol radiative forcing. In this study, an event of high pollution levels was investigated with an aim to understand drivers of single scattering albedo (SSA) alterations. Complex measurements of aerosol chemical composition and main parameters (aerosol light scattering and absorption coefficients, absorption and scattering Angstrom exponents, symmetry parameter, aging, size, aerosol liquid water content and other) were performed in urban environment in Lithuania during residential heating season (from 10th October to 1st November 2014). Residential heating and traffic related organic aerosols (BBOA and POA, respectively) exhibited higher mass absorption cross-section values (MAC) (1.14 m2 g-1 and 1.68 m2 g-1, respectively) showing an important impact on total light absorption by aerosols. During high pollution event, PM10 mass concentration reached up to 156.7 µg m-3. An evident input of not photochemically aged and small particles was observed. In addition, a significant increase in both black and brown carbon (BC and BrC, respectively) levels was observed, which was associated with accumulation of anthropogenic primary aerosol. During the event, alterations of SSA were mostly associated with changes in PM1/BC ratio. |

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| Madeleine | Petersson Sjögren | Lund University | Jakob Löndahl | Seasonal patterns in atmospheric fluorescent bioaerosol concentrations measured during 16 months in a forest in the south of Sweden | Biological aerosol particles are essential for the spread of organisms and are important agents for atmospheric processes, such as cloud formation and precipitation. However, the atmospheric abundance and size distribution of biological particles are largely unknown. In this study we have used a Laser-Induced Fluorescence instrument to measure fluorescent biological aerosol particles (FBAPs) over 16 months in the south of Sweden. Averaged over the full campaign (October 2020 – Jan 2022), the mean FBAP number concentration was 0.01 cm ⁻³ , corresponding to ~2% of the total coarse particle number concentration. The FBAP abundance depends strongly on season, with the highest FBAP concentration measured in the summer (0.90 cm ⁻³). Our measurements show that the FBAP concentrations are dominated by particles with aerodynamic diameters ranging from 1 to 3 µm, suggesting that the main portion of atmospheric biological particles are due to single cells, bacteria, fungal spores and agglomerated bacteria. We also see that the FBAP concentrations are related to air temperature and relative humidity. To our knowledge this is the first exploratory study reporting continuous long-term online measurements of bioaerosols in northern Europe. Our results confirm that bioaerosols account for a small proportion of coarse aerosol particle number and mass in boundary layer air. |
| Markus | Petters | NC State University/Aarhus University | Sabin Kasparoglu, Russell Perkins, Paul Ziemann, Paul DeMott, Sonia Kreidenweis, Zachary Finewax, Ben Deming, Marla DeVault, Markus Petters | Experimental determination of the relationship between organic aerosol viscosity and deposition mode ice nucleation at upper free tropospheric conditions | Atmospheric aerosols can exist in amorphous semi-solid or glassy phase states whose viscosity varies with atmospheric temperature and relative humidity. The particle phase state has important implications for intraparticle mixing time, chemical reactivity, and deposition mode ice nucleation in the upper free troposphere. For example, model single component glassy organic particles such as citric acid and sucrose have been shown to nucleate ice within the deposition ice nucleation regime (at RH lower than required for homogeneous freezing of dissolved aerosols) at temperatures below -40° C. However, studies of secondary organic aerosol freezing in this regime have provided mixed results, with numerous studies reporting difficulty in detecting freezing prior to homogeneous freezing onset. Constraining the phase state of the particles prior to freezing a critical piece of this puzzle. One method to study the viscosity and glass transition temperature of particles is to prepare coagulated dimer particles using the Dual Tandem DMA technique, followed by probing the thermodynamic states that induce the dimer particles to relax into spheres. Here we apply this method in conjunction with ice nucleation measurements for secondary organic aerosol between -40 °C and -65 °C. In all of our experiments the particles either froze consistent with the homogeneous freezing nucleation threshold indicating that they form an aqueous solution, or exceed water saturation indicating that they were solid and hydrophobic. Neither functional group composition nor close structural matches between known organic ice nuclei as precursor or formed products promoted freezing. We conclude by collating similar laboratory data across multiple studies. Combined these results suggest that complex secondary organic aerosols are unlikely to significantly contribute to the heterogeneous ice nucleating particle population in the upper free troposphere. |
| Sarah | Petters | Aarhus University | | Constraints on the role of Laplace pressure in multiphase reactions and viscosity of organic aerosols | Aerosol chemistry has broad relevance for climate and global public health. The role of interfacial phenomena in condensed-phase aerosol reactions remains poorly understood. In this work, liquid drop formalisms are coupled with high-pressure transition state theory to formulate an expression for predicting the size-dependence of aerosol reaction rates. Insights from high-pressure organic synthesis studies suggest that accretion and cyclization reactions are accelerated in 3–10-nm particles smaller than 10 nm. Reactions of peroxides, epoxides, furans, aldols, and carbonyls are predicted to be accelerated by up to tenfold. Effective rate enhancements are ranked as: cycloadditions >> aldol reactions > epoxide reactions > Baeyer-Villiger oxidation >> imidazole formation (which is inhibited). High-pressure changes to viscosity are predicted to further perturb nanoparticle reactions. Some reactions are enabled by the elevated pressure in particles. Results suggest that internal pressure is an important consideration in studies of chemical evolution of particles during new particle formation. |
| Marje | Prank | Finnish Meteorological Institute | Marje Prank, Tomi Raatikainen, Jaakko Ahola, Juha Tonttila, Harri Kokkola, Thomas Kühn, Sami Romakkaniemi | Investigating the role of sea-spray originating giant cloud condensation nuclei in drizzling marine stratocumulus using a large eddy simulator | Compared with fine particles that tend to extend the cloud lifetime, the effect of the coarse fraction is different - acting as giant cloud condensation nuclei (GCCN) they speed up the collision-coalescence process that can lead to earlier drizzle formation. Previous studies have shown that while marine stratocumulus can be highly sensitive to GCCN, the related uncertainties are large due to complex dependencies on the background CCN concentration and GCCN size distribution. We use the UCLALES-SALSA large eddy simulator that includes a detailed description of aerosols, cloud droplets, rain drops, and their interactions and parameterizations for sea spray emission to simulate the conditions of DYCOMS-II observational campaign. We aim to quantify the sensitivity of the marine low level stratiform clouds to the GCCN by varying the size range of emitted sea spray flux. Our results show that sea spray in the size range of 1-10 microns readily acts as GCCN and speeds up the drizzle onset noticeably when compared to control simulation without supermicron sea spray. Sea spray in the dry size range of 10-23 is also capable of reaching cloud level and has similar though smaller impact on drizzle onset. The effect of GCCN is sensitive to the fine mode aerosol concentration and assumptions regarding the hygroscopicity of the coarse particles. |

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| Mike | Priestley | Gothenburg University | Michael Priestley, Xiangrui Kong, Xiangyu Pei, Julia Hammes, Daniel Bäckström, Ravi K. Pathak, Jan B. C. Pettersson, Mattias Hallquist | Comparison of FIGAERO-ToF-CIMS derived volatility measurements of fresh and aged residential wood burning OVOCs | <p>One key aim of current aerosol research is to accurately represent the phase partitioning of semi-volatile organic compounds (SVOCs). Better representations of phase-resolved aerosol mass and composition would improve predictions of aerosol spatial and temporal variability and thus improve predictions of subsequent climate and health effects e.g. optical interactions and PM_{2.5} concentrations. Residential wood combustion (RWC) is a major source of organic aerosol, including semi-volatiles, at local and regional scales causing pollution in populated areas in Europe. Here we use a FIGAERO-ToF-CIMS to measure semi-volatiles emitted from a typical RWC stove. We derive partitioning coefficients for hundreds of compounds using two formulations of absorption partitioning theory and contrast these with two different composition-activity parameterisations.</p> <p>Correlations of molecular weight vs partitioning coefficients suggests partitioning theory is most appropriate for those emissions where the organic:elemental carbon ratio is high. Levoglucosan, a biomass-burning tracer, is the dominant compound regarding volatility distributions. Removing its influence reveals that the SVOC volatility distributions are similar for the two measurement derived results, which agree well with one of the parameterizations. However, once these distributions are weighted by mass, the agreements are lost. For the measurement derived distributions, this may be a consequence of high measurement uncertainties.</p> |
| Iida | Pullinen | University of Eastern Finland | Angela Buchholz, Arttu Ylisirniö, Lejsh Vettikkat, Sami Romakkaniemi, Ari Leskinen, Siegfried Schobesberger, Anneli Virtanen | PMF ANALYSIS OF FIGAERO-CIMS DATA: EFFECT OF THE AIRMASS ORIGIN AND LOCAL EMISSIONS ON DETAILED SOA PARTICLE COMPOSITION | <p>As a part of the EU funded FORCes project, measurements took place at the semi-urban Puijo research station SMEAR IV, situated in central eastern Finland. The measurement platform is situated at the top of Puijo tower, and due to its elevation, the measurement station is frequently in-cloud, providing an opportunity to explore cloud-aerosol interactions directly within cloud. The chemical composition of organic atmospheric aerosol was measured for several weeks during autumn 2020. Several aerosol properties were measured simultaneously: A FIGAERO-CIMS enabled a detailed analysis of the molecular composition and properties of the measured SOA. A DMPS/SMPS system was present for particle number and mass concentration information. An ACSM provided information of the general SOA mass composition and oxidation states/oxidation properties. The gas phase composition was measured by a VOCUS. Additionally, meteorological parameters were monitored, and trace gases such as ozone, SO₂, NO_x concentrations were measured.</p> <p>Using FIGAERO-CIMS data, we analysed how the SOA properties (such as composition and volatility) changed over the measurement period. Ambient datasets are by their nature complex and have many factors contributing to the properties of the measured aerosol bulk, e.g. the origin of the aerosols, its age and the processing it has experienced, and possible local versus long distance sources. For these reasons a change in the wind direction can in some cases lead to large changes in the observed particle properties, and it can be difficult to distinguish different processes in play. To detangle this complex dataset, a PMF analysis was conducted where the overall longer timeseries data of the chemical composition of the SOA is combined with the additional compound volatility information for each individual sample provided by the instrument to separate different ambient factors of the measured data.</p> <p>The duration of the measurement campaign was approximately 2 months, from which a period of three weeks was selected for a closer analysis. This period includes a so-called plume period, with clearly elevated aerosol concentration of up to ~15 µg m⁻³ compared to the average of ~1.0 µg m⁻³ during the rest of the campaign (based on SMPS data, excluding this high mass period). The elevated SOA concentrations and much larger particle sizes were linked to air masses transporting aerosols from forest fires in (south)eastern Europe to our measurement site. It was followed by a period of varying SOA loadings, providing contrast and the lower SOA loadings expected at a (semi-)rural measurement station. The preliminary PMF analysis showed that the plume period was clearly distinguishable from the other times. The results from the PMF analysis are compared and combined with other analysis methods.</p> |
| Tomi | Raatikainen | Finnish Meteorological Institute | Tomi Raatikainen, Marje Prank, Jaakko Ahola, Harri Kokkola, Juha Tonttila, Sami Romakkaniemi | Cloud effects of marine ice-nucleating particles | <p>Some recent studies have concluded that ice-nucleating particles (INPs) emitted as sea-spray aerosol have significant impacts on shallow clouds over remote regions such as the Southern Ocean. To estimate the potential impacts of marine INPs on shallow mixed-phase clouds, we made a series of Large Eddy Simulations (LES) with different background dust INP concentrations and with marine INP emissions either on or off (Raatikainen et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-537, in review, 2021). The model in these simulations is UCLALES-SALSA, which was recently extended for mixed-phase clouds (Ahola et al., Atmos. Chem. Phys., 20, 11639-11654, 2020). The simulations showed that marine INPs are transported efficiently from the sea-surface up to the cloud layer. As expected, marine INPs have the largest impact on clouds when the background INP concentration is low. The detailed simulations also showed that ice nucleation took place most often in updraught regions. The updraughts were also enriched in INPs originating from the previously sublimated ice crystals.</p> |

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| Maher | Sahyoun | Department of Environmental Science, Aarhus University | Maher Sahyoun, Kostas Tsigaridis, Tina Santl-Temkiv, Ulas Im | Introducing Primary Biological Aerosol Particles in GISS-E2.1 Earth system model | <p>Primary biological aerosol particles (PBAPs) are present globally, contributing to the total observed aerosol loads. Yet, PBAPs likely form a smaller fraction of the total aerosol budget compared to other types of particles such as dust. According to the IPCC AR5 report, the terrestrial emission flux of PBAPs is highly uncertain and was estimated within the range of 50-1000 Tg/yr. Burrows et al. (2009) estimated the global emissions of bacteria-containing particles to range between 0.4 to 1.8 Tg/yr, with a median of 0.74 Tg/yr. However, PBAPs comprise a large fraction of the submicron particles > 0.2 μm in the middle to the upper troposphere and they can be dispersed to distant locations and high altitudes from their source regions. PBAPs have the potential to play a key role in cloud formation by acting as cloud condensation nuclei (CCN), and ice nucleating particles (INP) active at high sub-zero temperatures; potentially impacting the Earth's hydrological cycle and climate.</p> <p>Recent observations suggest that the PBAPs' concentrations have likely been underestimated in global modeling studies (summarized in Huang et al., 2021). For example, the fragmented biological particles and microbial exudates still cannot be detected with many commonly used techniques and, therefore they were not accounted for in the previous global modeling studies. Other recent studies presented a novel secondary biological aerosol production. Moreover, observations revealed that biological INPs from marine surfaces may be of higher importance than what has previously been considered in modeling studies. PBAPs' emission flux is therefore not yet well constrained, and the uncertainty in their emission estimation remains unresolved and requires deeper investigation. Consequently, the climatic impacts and feedbacks of PBAPs remain highly uncertain.</p> <p>In this study, we build and integrate for the first time a new emission model for PBAPs in the GISS-E2.1 Earth system model in order to calculate the total emission flux of PBAPs from terrestrial and marine surfaces into the atmosphere and estimate their transport and sinks. In this model, we consider different types of PBAPs, i.e., bacteria and fungal spores. For bacteria, we used the estimated flux rates from Burrows et al. (2009) for different ecosystems. In a later step, we plan to update those values for each ecosystem using recent observations, especially over the marine areas and land-ice. For fungal spores, we used the parameterization of Janssen et al. (2021).</p> <p>We present preliminary results of the emission fluxes, burdens, concentrations, lifetime, and direct radiative forcing due to aerosol-radiation interactions of PBAPs and validate them using previous studies, e.g., Burrows et al. (2009) and Hoose et al. (2010).</p> |
| Prasenjit | Seal | Tampere University | Olga Garmash, Matti P Rissanen | Mechanistic Study of the oxidation of Naphthalene by OH radical: A computational perspective | <p>Anthropogenic volatile organic compounds (AVOCs) often dominate the urban atmosphere and consist to a large extent, aromatic hydrocarbons. These aromatics are important precursors for the formation of secondary organic aerosols. Naphthalenes are the most abundant polycyclic aromatic hydrocarbons in polluted urban areas and is reactive in ambient atmospheres. In the present work, we proposed a mechanistic pathway for the oxidation of naphthalene by OH radicals, which leads to the formation of highly oxygenated molecules (HOMs). The mechanism further is justified by experimental observations. The electronic structure calculations were performed with high-level quantum chemical methods. We also estimated rate coefficients of some crucial steps of the mechanistic pathway using master-equation simulations.</p> |
| Jane | Skonager | Aarhus University | Merete Bilde | Phase state transition of Δ^3 -carene derived secondary organic aerosols | <p>The viscosity of secondary organic aerosol (SOA) can vary with relative humidity and temperature. It also affects particle properties such as particle size distribution and mass concentration. Furthermore, the diffusivity of organic molecules, water and oxidants in the particles is also affected by the viscosity, which can then alter the equilibrium time scales, particle lifetime and gas-particle partitioning.</p> <p>The phase state of aerosols is described by the dynamic viscosity. Multiple studies have shown that the phase state of atmospheric particles can vary from liquid-like states to amorphous solid states, i.e., glassy states. The phase state of particles affects the cloud-forming properties of the aerosol since aerosols in a glassy state can act as ice nuclei and impact cirrus cloud formation processes.</p> <p>The viscosity of SOA is difficult to measure or probe experimentally. In this study, the viscosity of Δ^3-carene derived SOA is explored specifically by searching for indications of a phase transition between a solid glassy state and a semi-solid or liquid state. The temperature where this transition occurs, is called the glass transition temperature. The glass transition is not a classical transition with an exact transition temperature, but rather a transition that occurs over a temperature interval. Nonetheless, it is very useful to define a glass transition temperature, T_g, that represents this temperature interval, and the particle phase state can then be related to the glass transition temperature.</p> <p>We present preliminary results of experiments performed in the AURA atmospheric simulation chamber at Aarhus University. SOA particles are formed from dark ozonolysis of Δ^3-carene, and temperature induced phase transitions are investigated in temperature ramp experiments. Changes in aerosol size distributions are measured with a Scanning Mobility Particle Sizer system. The chemical composition is measured using an Aerosol Mass Spectrometer.</p> <p>The results are compared to results from similar experiments done in the AURA atmospheric simulation chamber studying ozonolysis of α-pinene. α-pinene and Δ^3-carene are structurally similar monoterpenes, but studies indicate that the particle formation from ozonolysis of α-pinene and Δ^3-carene is different.</p> |

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| Gerhard | Steiner | GRIMM Aerosol Technik | Markus Pesch, Friedhelm Schneider, Torsten Bayer, Michael Schüler, Katharina Roloff and Gerhard Steiner | Continuous particle number concentration determination of ambient air in measurement networks | <p>Recently WHO published new Global Air Quality Guidelines (WHO, 2021), once more stressing the danger of air pollution on human health, and addressing this, for the first time, also to the number of ultrafine particles (UFP, particles < 0.1 µm) in the ambient air. No definitive air quality guidelines are expressed for UFP yet, however, WHO highlights best practices and the importance of the continuous measurement of ultrafine particles.</p> <p>Furthermore, the European CEN/TS 16976:2016 guideline expands the need to measure ultrafine particles in the ambient air also to address the impact of UFP on the global climate. This guideline also underlines the need to determine the particle number (PN) of the ambient aerosol to supplement commonly applied measurement procedures of determining particulate mass fractions such as PM10 or PM2.5, as mass is not a feasible metric for ultrafine aerosols.</p> <p>This CEN/TS aims to harmonize currently available measurement techniques for the determination of the particle number concentration in the ambient air, thereby defining instrument performance characteristics such as overall instrument counting efficiency, D50/D90 diameters and specifying the range of measurement uncertainties in instrument calibrations. Here, we are reporting preliminary results of two 1-month measurement campaigns in Halle (Saale), Germany and Berlin, Germany, carried out in collaboration with the authorities operating the local air quality monitoring networks. In both measurement campaigns, instruments were deployed at existing measurement stations close to heavily frequented streets and at rather remote background locations and operated with high temporal resolution.</p> <p>For the determination of the particle number of ultrafine aerosols, we were using the GRIMM condensation particle counters EDM 465 and CPC 5421, specifically designed for 24/7 use and mounted in dedicated weatherproof and climate controlled outdoor shelters. Both instrument models were calibrated in accordance to CEN/TS 16976:2016 and were using sample collecting probes with integrated membrane dryers. In addition to the UFP measurements, PM10 and PM2.5 values were determined with GRIMM EDM 180 optical aerosol spectrometers.</p> <p>We will give details on the used measurement instruments, describe the measurement sites, and will give an outlook on possible data analysis using GRIMM aerosol monitors for long term measurements.</p> <p>Acknowledgement: We would like to thank the Senate Department for the Environment, Urban Mobility, Consumer Protection and Climate Action of the City of Berlin for their collaboration and the access to their measurement sites within the Berlin air quality network BLUME.</p> |
| Sara | Thuresson | Lund University | Carl-Johan Fraenkel, Patrik Medstrand, Malin Alsvéd, Jakob Löndahl | SARS-CoV-2 in size-fractionated aerosols from hospital corridors and relations to the indoor environment | <p>Although most hospital wards treating patients with infectious diseases have isolation rooms with airlocks and regulated pressure systems, there is a risk that virus might be present in the air of corridors. In the corridors, less personal protective measures are in place than in patient rooms, posing a risk of infection for the hospital staff. Airborne virus might also be emitted by pre- or asymptomatic staff.</p> <p>The aim of the current study was to evaluate the presence of airborne virus, specifically SARS-CoV-2, in infection ward corridors, and the size distribution of the virus-containing aerosols. Associations between virus presence and relative humidity and/or temperature in the facilities was also investigated.</p> <p>Aerosol particles were collected in hospital corridors from March 2020 to April 2021 at infectious disease wards in Southern Sweden. Collection was performed using an 8-stage cascade impactor (Next Generation Impactor, Copley Scientific, UK) operating at a flowrate of 60 L min⁻¹ for 12 hours a day, 7 days a week. The impactor plates were exchanged every 7 days. After collection, each impactor stage was swabbed with a wetted nylon swab (Copan Scientific) and stored in universal transport media in -80 °C until analysis with real time reverse transcription polymerase chain reaction (RT-qPCR) for detection of SARS-CoV-2 RNA. The size fractions collected were: >8.1 µm, 4.5–8.1 µm, 2.9–4.5 µm, 1.7–2.9 µm, 0.9–1.7 µm, 0.6–0.9 µm, 0.3–0.6 µm, and 0.1–0.3 µm.</p> <p>Indoor temperature, relative humidity and CO₂ concentration was recorded with a CL-11 or CP-11 multiple parameter meter (Rotronic, Germany). Recording was done 24 hours a day, 7 days a week, during the entire sampling period.</p> <p>So far, only a small fraction of the collected samples (48 of 816) have been analysed by RT-qPCR. The majority of these are negative for SARS-CoV-2. Temperature measurements showed a mean corridor temperature of 23.4°C (SD: 0.38). Relative humidity varied between 6 and 67 % (mean: 28.4, SD: 10.6).</p> <p>Results are expected to increase our understanding about virus presence in hospital corridors, and what aerosol particle sizes that contain SARS-CoV-2. Size information is important for understanding the particle origin, transmission patterns indoors, and where these particles deposit in the respiratory system when inhaled. Possible relations to temperature and relative humidity could improve mitigation strategies related to controlling the indoor environment.</p> |
| Valtteri | Tikkanen | Institute for Atmospheric and Earth System Research/Physics, Faculty of Science, University of Helsinki, P.O. Box 64, FI-00014, Finland | Bernhard Reischl, Hanna Vehkamäki, Roope Halonen | Homogeneous nucleation in the gas phase is driven by cool subcritical clusters | <p>Classical nucleation theory assumes that clusters nucleating in the gas phase are in thermal equilibrium with their surroundings. However, a significant amount of energy is released upon cluster formation, and how this affects nucleation rates or cluster temperatures remains controversial. Molecular dynamics simulations of homogeneous nonisothermal nucleation of a supersaturated vapor in the presence of carrier gas allow us to study the real-time dynamics and energetics of the system. We find that nucleation rates are suppressed by two orders of magnitude at most, despite substantial release of latent heat. Our simulations further reveal that while the temperatures of the entire cluster size populations are elevated, the temperatures of the specific clusters driving the nucleation flux evolve from cold to hot when growing from subcritical to supercritical sizes. Our findings provide unprecedented insight into realistic nucleation events and allow us to directly assess earlier theoretical considerations of nonisothermal nucleation.</p> |

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| Velle | Toll | University of Tartu | Velle Toll, Jorma Rahu, Heido Trofimov | Natural experiments help to understand aerosol-cloud interactions | Anthropogenic aerosols offset an unknown fraction of greenhouse gas warming, whereas the aerosol impact on clouds is the most uncertain driver of anthropogenic climate change. Clouds polluted by aerosols from localized pollution sources like ships, volcanoes, wildfires, and various industrial sources - such as oil refineries, smelters, coal-fired power plants, and cities serve as natural experiments of aerosol-cloud interactions (Toll et al. 2019; Nature, https://doi.org/10.1038/s41586-019-1423-9). However, ship-track-like polluted cloud tracks do not always form when suitable pristine clouds are present at a pollution source. In some cases, clouds become polluted just after sunrise, possibly indicating the importance of photochemistry in aerosol formation. |
| Zhuyun | Ye | Aarhus University | Camilla Geels, Jørgen Brandt, Lise Marie Frohn, Jesper Heile Christensen, Jeremy D. Silver, Arjo Segers, Kaj Mantzius Hansen | Three-dimensional variational data assimilation of particulate matters with the Danish Eulerian Hemispheric Model | In the frame of the Copernicus Atmosphere Monitoring Service (CAMS), we have coupled a three-dimensional variational data assimilation tool to the Danish Eulerian Hemispheric Model (DEHM) for near real time (NRT) operational analysis services of Europe. In the current study, we assessed the performance of the data assimilation scheme on PM2.5 and PM10 simulations during summer and winter periods. The hourly ground measurements used in the data assimilation and model evaluation are gathered from the centralized AirBase database maintained by the European Environment Agency (EEA). The simulations of both PM2.5 and PM10 were effectively improved with data assimilation in both seasons, and more significant improvements were shown for PM2.5 in summer. Sensitivity simulations were also performed to test the selections of observational stations. Using observations from both rural and urban sites showed better simulation performances in various regions than using only rural observations. |
| Iiona | Ylivinkka | University of Helsinki / INAR | Iiona Ylivinkka, Santeri Kaupinmäki, Meri Virman, Maija Peltola, Ditte Taipale, Tuomo Nieminen, Tuukka Petäjä, Veli-Matti Kerminen, Markku Kulmala and Ekaterina Ezhova | Analysis of the effects of cloud types on aerosol-cloud-radiation interactions at SMEAR II | Clouds and aerosol particles can help in mitigating climate change via feedback mechanisms between atmosphere and ecosystem. Previous studies have shown that photosynthesis of a forest ecosystem can be fertilized due to rising CO2 concentration and air temperature, leading to enhanced emissions of biogenic volatile organic compounds, some of which can contribute to formation of secondary organic aerosol after being oxidized in the atmosphere. The formed aerosol particles may act as cloud condensation nuclei, altering thus cloud properties. Clouds and aerosol particles are both efficient in backscattering solar radiation, thereby cooling the climate, but they also increase the fraction of diffuse radiation, which have been shown to further benefit photosynthesis on ecosystem scale. However, large uncertainty is related to these feedback mechanisms because the properties of aerosol particles and different cloud types vary greatly, affecting thus the impacts they have on climate. To study the effects of cloud types on various boundary layer processes, including for example new particle formation (NPF) and photosynthesis, we developed a cloud classification algorithm, applying global radiation and cloud base height measurements from the SMEAR II (Station for Measuring Ecosystem-Atmosphere Relations) measurement site in Hyttälä, Finland. From measured and modeled global radiation two parameters, transmittance and patchiness, can be derived for cloud type separation. Transmittance describes the amount of radiation coming through the clouds, and patchiness indicates the variability in the cloud layer. Due to possible bias in global radiation measurements at large solar zenith angles (SZA), the algorithm is valid when $SZA < 70^\circ$. At SMEAR II the SZA limit leads to exclusion of wintertime data, meaning that the results by the algorithm are valid from end of February to middle of October. The algorithm can distinguish stratus, stratocumulus, cumulus, nimbostratus, altostratus + altocumulus, cirrus + cirrostratus + cirrocumulus, and clear sky + cirrus clouds, categorize multilayered cloud cases, and cases when the cloud type could not be classified. The overall performance of the algorithm is nearly 70 %. When studying cloud types at 9:00–12:00 (UTC+2) on NPF event and non-event days we noticed that on most cases (41 %) NPF events occur on clear-sky conditions, but NPF is also common in the presence of clouds with high patchiness and transmittance. For example cumulus clouds were present in 27 % of the cases. Further analysis is required to determine what is the contribution of clouds on NPF and the processes described above. For these purposes, the developed cloud algorithm is a needed new tool, promoting holistic studies on aerosol-cloud-radiation interactions. |
| Putian | Zhou | University of Helsinki | Zhengyao Lu, Jukka-Pekka Keskinen, Jianpu Bian, Jason Zhang, Qiang Li, Qiong Zhang, Risto Makkonen | Simulated SOA over Sahara in Mid-Holocene with prescribed vegetation cover and BVOC emissions | A Green Sahara, instead of present desert Sahara, existed in the North-Africa during the early- to mid-Holocene (11,000 to 5,000 years before present) as suggested by comprehensive paleoproxy data and studies. In this study, we applied the global transport model TM5 to simulate the global dust load concentrations and the formation of secondary organic aerosols (SOAs) during mid Holocene (6000 year before present) period. With reconstructed mid-Holocene vegetation covers, as well as isoprene and monoterpene emissions, we found that compared to per-industrial period, the SOA load is 1.2 mg m ⁻² higher and CCN0.2 (cloud condensation nuclei at 0.2% supersaturation) concentration increased by 10-25% over North Africa during mid-Holocene period. This may increase the precipitation indicating a positive feedback for northward shift of vegetation in Sahara region. |

Phase behavior in organic aerosol mixtures is driven by difference in oxygen-to-carbon ratio of components

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Atmospheric aerosols play an important role for air quality and climate. Primary organic aerosol (POA) and secondary organic aerosol (SOA) make up ~20-90% of the mass fraction of tropospheric aerosol. POA denotes aerosols that are directly emitted into the atmosphere as particles, while SOA mostly forms from oxidation of precursors, followed by gas-particle conversion of the oxidation products. Representation of SOA formation in atmospheric models and prediction of impacts on air quality and climate requires knowledge of the phase behavior (the number and types of phases) in internal mixtures containing SOA together with other aerosol types. For instance, in the case of POA+SOA particles, it is often assumed that SOA formation is enhanced 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 particles. The presence of POA will have a smaller effect on the formation of SOA mass in cases of phase separated (two-phase) POA+SOA particles.

Here, using fluorescence microscopy, we observed the phase behavior of individual particles containing mixtures of different aerosol materials. We show that in POA+SOA and SOA+SOA particles, the difference in the O/C ratio (a proxy for difference in polarity) between the components of a mixture is a good predictor, whether one- or two-phase particles form. When adding secondary inorganic aerosol (SIA), another important atmospheric particle type, we find that three phases can often co-exist in POA+SOA+SIA particles. Our results have important implications for air pollution policies being considered to limit SOA formation in urban environments.

How does High Latitude Dust affect the air quality and climate in the Arctic and Antarctica?

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High Latitude Dust (HLD) contributes 5% to the global dust budget and active HLD sources cover > 500,000 km². Potential areas with high HLD emission in Iceland, Canada, Alaska, Svalbard, Greenland, North Eurasia, Patagonia, Antarctica, and New Zealand are calculated to cover >1,670, 000 km². Iceland is the largest European desert with 44,000 km². Recent studies have shown that Icelandic dust travelled about 2,000 km to Svalbard, about 3,500 km to Balkan Peninsula and it can reach Scandinavia at least 10 times per year. It estimated that about 7% of Icelandic dust can reach the high Arctic (N>80°). HLD was recognized as an important climate driver in Polar Regions in the IPCC Special Report on the Ocean and Cryosphere in a Changing Climate in 2019.

In situ dust measurements conducted in Arctic deserts of Iceland and Antarctic deserts of Eastern Antarctic Peninsula revealed some of the most severe dust storms in terms of particulate matter (PM) concentrations, resulting in impaired air quality. While one-minute PM₁₀ concentrations in Iceland exceeded 50,000 µg m⁻³, ten-min PM₁₀ means in James Ross Island, Antarctica exceeded 120 µg m⁻³. Icelandic dust has impacts on atmosphere, cryosphere, marine and terrestrial environments. It decreases albedo of both glacial ice/snow as well as mixed phase clouds via reduction in supercooled water content. There is also an evidence that volcanic dust particles scavenge efficiently SO₂ and NO₂ to form sulphites/sulfates and nitrous acid.

Large HLD field campaign has been organized as well as first HLD operational dust forecast for Icelandic dust is available at the World Meteorological Organization Sand/Dust Storm Warning Advisory and Assessment System (WMO SDS-WAS) at <https://sds-was.aemet.es/forecast-products/dust-forecasts/icelandic-dust-forecast>. HLD research community is growing and Icelandic Aerosol and Dust Association (IceDust) has 100 members from 48 institutions in 18 countries (<https://icedustblog.wordpress.com>, including references for this abstract).