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Atmospheric surfaces

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It is with pleasure that we introduce this themed issue of *Environmental Science: Processes & Impacts* on the topic of "Atmospheric Surfaces". Atmospheric surfaces are ubiquitous on all scales ranging from nanometer-sized surfaces, such as the surfaces of particles and cloud nuclei, to global scale including marine and terrestrial surfaces. Of particular interest in this issue are:

- air–water interactions – from cloud condensation nuclei to marine surfaces
- air–ice interactions – atmosphere–cryosphere interactions and ice nuclei processes
- reactive surface processes – such as reactions on aerosol surfaces or sea surfaces
- air–soil interactions – *e.g.* exchange of gases

Molecular scale transport and interactions on atmospheric surfaces have tremendous influence on processes affecting air quality and climate. There is a need for research on these topics to obtain detailed understanding enabling accurate parameterizations for implementation in local, regional and global models. The purpose of this themed issue is to provide an overview of recent research results and the state-of-the-art in this broad field.

Several of the papers in this issue focus on method development in different areas relating to the measurement of properties of aerosol particles. Gorkowski *et al.* (DOI: 10.1039/C8EM00166A) describe a new algorithm for the measurement of the size and evolution of core–shell particles using the whispering gallery modes seen in Raman microscopy of levitated particles. Their method reduces the number of parameters, which allows for faster computing times, and thereby allows for the study of particle evolution over several hours with 0.5 Hz measurements. Tirella *et al.* (DOI: 10.1039/C8EM00276B) have extended surface enhanced Raman spectroscopy (SERS) to 150–800 nm particles using silver foil substrates. They observe enhancements in signal-to-noise of 10^2 compared to traditional Raman microscopy. In addition, they probed the composition of a 150 nm particle, which is below the diffraction limit. Such a technique has promise for the identification of the composition of individual particles, which could be a complement to single particle mass spectrometry. Finally, Cui *et al.* (DOI: 10.1039/C8EM00308D) describe hydrophilic interaction liquid chromatography (HILIC)/ESI-HR-quadrupole time-of-flight mass spectrometry (QTOFMS) which can chromatographically resolve and measure water soluble IEPOX-derived secondary organic aerosol (SOA) constituents. IEPOX is a major product of isoprene oxidation, which subsequently produces water soluble 2-methyltetrols and methyltetrol sulfates. Previous techniques used to measure these had poor separation and/or poor efficiency for polar compounds. This new method will allow for more accurate estimates for these water-soluble components of SOA.

Addressing the interactions between aerosol particles and water, Lin *et al.* (DOI: 10.1039/C8EM00345A) describe a new model to investigate the role of surface-active species in cloud condensation nuclei (CCN) activation. Their method uses their previously developed monolayer model combined with Köhler theory to describe this process. While their model is consistent with Gibbs thermodynamics, it requires many fewer material-specific inputs. As a result, the monolayer model can be more easily applied to complex systems of atmospheric relevance.

Processes of particular relevance in the marine boundary layer are investigated in the paper by Li *et al.* (DOI: 10.1039/C8EM00419F), who studied mixed Langmuir monolayers of cholesterol with stearic acid and oleic acid at the air–seawater surface. These oleic acid/cholesterol monolayers were found to be thermodynamically stable, while the ordering of fatty acid monolayers at

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the air–seawater surface was changed by mixing with cholesterol. The results are relevant for understanding of organic coating of sea salt aerosol.

Two papers address air–ice interactions, specifically focusing on the ice nucleation of aerosol particles. Losey *et al.* (DOI: 10.1039/C8EM00319J) performed chemical characterization and immersion freezing studies of fly ash before and after treatment with sulfuric acid. The behaviour of the particles to ice nucleation depends on the composition of the fly ash, specifically whether it was rich in calcium, iron, or both, with some samples showing increased and others showing decreased activity towards ice nucleation when treated with acid. The significance of this study is the variation in the behaviour of a sample we typically group as a single aerosol type, "fly ash". DeMott *et al.* (DOI: 10.1039/C8EM00386F) performed a study on the ice nucleation of fatty acids with relevance to marine chemistry. They were unable to observe ice nucleation on aqueous particles with fatty acids at the surface unless a crystalline phase was present. Compression of the monolayer did not affect the ice nucleation activity. This is in contrast to fatty alcohols, which can nucleate ice at temperatures approximately 20 °C warmer than these fatty acids.

Two papers address reactive processes of aerosol particles. Vander Wall *et al.* (DOI: 10.1039/C8EM00348C) measured the uptake of three unreactive organonitrates onto hydrophobic and hydrophilic organic films. These studies are important for understanding the incorporation of gases into particles, which can lead to reactions and particle growth. They find that uptake and partitioning do not follow the same trends, indicating that gas–surface interactions differ from those in the bulk. Glasius *et al.* (DOI: 10.1039/C8EM00413G) investigated the organosulfates formed from the reaction of organic compounds, such as gasphase isoprene oxidation products, and acidic sulfate aerosol. They detected organosulfates downwind of the city of Manaus in the Amazon rainforest in Brazil during GoAmazon 2014/15. The isoprene derived organosulfates contributed considerably to the total sulfate aerosol resulting in an average organosulfate to sulfate ratio much higher than observed elsewhere.

The final paper investigates air–soil interactions. Mielnik *et al.* (DOI: 10.1039/C8EM00356D) investigated the emission of formic and acetic acid from two Colorado soils using a high-resolution time-of-flight chemical ionization mass spectrometer. While acid emissions do not correlate directly with CO_2 fluxes, they increase exponentially with temperature, and the emission of acetic acid increases with moisture content. There is a large missing source of formic acid in global budgets; while soils are not likely to be this missing source, the variability in emissions is more than what is currently accounted for in models.

We hope that you, the readership of *Environmental Science: Processes & Impacts*, enjoy this collection of papers on Atmospheric Surfaces. We are thankful to the contributors of the articles and to the anonymous reviewers who help bring these works to publication. In addition, we gratefully acknowledge the editors and staff at the Royal Society of Chemistry who helped us put this themed issue together. And to you, the reader, we hope that this collection inspires you to further explore this timely scientific topic.