Insights into key processes governing atmospheric aerosol loadings and their interactions with clouds

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Abstract

Aerosol particles are ubiquitous in the atmosphere and an essential part of the atmospheric radiation balance regulating the Earth's temperature. Aerosol-cloud interaction still remains the largest single uncertainty in future climate projections. In addition, aerosols are also responsible for air pollution, causing severe health effects. With various origins and short atmospheric lifetimes, aerosols are unevenly distributed in the atmosphere, making simulations of air pollution and future climate scenarios challenging. This thesis aims to improve the understanding of the physical and chemical processes that govern aerosol concentration in the atmosphere, using both field as well as laboratory experiments.

Field measurements were performed at a remote station at Mt Åreskutan, central Sweden. Located at 1250 m a.s.l. the station is frequently covered by clouds, allowing for in-cloud measurements. Aerosol particle size distribution measurements revealed a shift towards smaller diameters in the ambient aerosol size distribution after the station had been within a cloud. This is a result of the larger (> 60 nm) particles being more effectively scavenged by clouds as compared with the smaller end of the size distribution. Chemical analysis revealed a similar composition of the cloud water as the particulate matter, suggesting that cloud droplet activation at Mt Åreskutan is primarily dependent on particle size, and the aerosol population to have been internally mixed. Similarly, measurement of hygroscopicity and volatility revealed similar water-solubility and evaporation behaviour for the ambient aerosols and cloud residuals, with the organic fraction representative of aged boreal secondary organic aerosol (SOA) and showing no signs of significant aqueous phase processing.

The NArVE laboratory campaign took place in an atmospheric simulation chamber at Paul Scherrer Institute, Switzerland. The experiments traced nitrate-induced SOA formation and ageing of three biogenic precursors, namely α-pinene, isoprene, and β-caryophyllene, using mass spectrometric techniques and evaporation measurements. The volatility of α-pinene SOA from nitrate oxidation was found to be higher than the corresponding ozonolysis products. The nitrate oxidation of isoprene resulted in species with similar volatility to α-pinene, while the β-caryophyllene system produced lower volatility compounds then the other two precursors. Quantitative comparison of the volatility measurements to commonly-used theoretical parameterizations revealed the need for further studies of the impact of the nitrate functional group on molecular volatility. Dark ageing of α-pinene was found to mainly occur through particle phase oxidation forming less volatile species. During the photolysis related to sunrise the molecular composition changed towards more volatile species, while no significant evaporation could be observed for the α-pinene and isoprene systems.

A common theme in all these studies was investigating the level of detail needed to theoretically describe the observations. We found that while simple approximations (such as internal mixing and size-independent chemical composition of the particles) are often sufficient to capture trends in atmospheric aerosol properties, more research on (1) the processes taking place on shorter time- and smaller size scales than investigated here and (2) the effects of nitrate group on molecular volatility are warranted.

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