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Ageing of sea spray aerosols: significance for hygroscopic growth and cloud droplet activation

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Sea spray aerosols (SSA) are continuously emitted into the atmosphere from the ocean and play a significant role in the planetary energy budget by interacting directly with solar radiation and by affecting the formation and lifetime of clouds. This has a major effect on climate since 70% of the planet is covered by ocean, representing the largest single source of aerosol mass to the atmosphere. The atmospheric lifetime of SSA varies from seconds to weeks. During this time, SSA is exposed to oxidants, condensable vapors, sunlight and changing conditions of relative humidity and temperature, which induce so-called ageing effects. Hence, the properties of SSA may be significantly altered as a result of aging, which leads to changes in their role for climate.

The hygroscopic behavior of SSA at relative humidities (RH) below 100%, influences the type and magnitude of the aerosol-radiation interactions as it changes the size and chemical composition and thereby the optical properties of the aerosol particles. The water uptake can also influence the particles' efficiency to act as cloud condensation nuclei (CCN) at supersaturated conditions. Commonly, SSA is believed to behave in a similar way as sodium chloride (NaCl), which is the major chemical component of SSA particles overall. Studies with more complex salt mixtures have however pointed to clear differences regarding the hygroscopicity (Zieger et al., 2017) and underline the importance of other compounds present in SSA. Previous laboratory studies have investigated reactions of aqueous NaCl with ozone and the presence of UV-light, hereby mimicking ageing conditions occurring in the marine boundary layer (Laskin et al., 2003). These experiments demonstrated changes in the chemical and physical properties of the particles and hence the authors hypothesize a possible effect on the hygroscopic and cloud activation potential.

In this study, we investigate the hygroscopicity and cloud forming potential of SSA after atmospheric ageing processes. The experiments are performed within the constrained environment of the Aarhus University Research on Aerosol smog chamber which allows for a controlled temperature regulation between 257 and 299 K. In a first step pure NaCl particles were studied and the results were then compared to more complex inorganic mixtures, that better represent the atmospheric SSA. Additionally, two generation techniques were employed: an atomizer and a sea spray chamber imitating the SSA production by breaking waves. The hygroscopic growth was measured with a humidified tandem differential mobility analyzer while a cloud condensation nucleus counter was used to investigate the cloud droplet activation of the fresh and aged particles. Besides, microscopy techniques were utilized to study the chemical composition of the particles. We will present first results of ageing experiments, where NaCl undergoes oxidation due to gases and UV-radiation as present in the atmosphere. We will show that only deliquesced NaCl particles lead to chemical reactions influencing the hygroscopic and CCN-activity and that the results strongly depend on the pre-treatment of the aerosol particles.

Laskin, A. et al. (2003), Science, 301: 340-344.
Zieger, P. et al. (2017), Nature Comm., 8: 15883.