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Photochemistry of atmospheric organic aerosols: Can chemical mechanisms be linked to aerosol-cloud interactions?

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Abstract

An organic aerosol particle has a lifetime of approximately 1 week in the atmosphere during which it will be exposed to sunlight. However, the effect of photochemistry on the propensity of organic matter to participate in the initial cloud-forming steps is difficult to predict. In this presentation, I will discuss the molecular scale effect of photochemical exposure of naturally occurring dissolved organic matter from rivers and from wood burning on its cloud condensation nuclei (CCN) and ice nucleation (IN) activity. Our group has found that photochemical processing, equivalent to 4 days in the atmosphere, of organic matter increases its ability to form cloud droplets but decreases its ability to form ice crystals. Specifically, the ice nucleation activity of photooxidized organic matter can require up to 4 °C colder temperatures for 50 % of the droplets to activate as ice crystals under immersion freezing conditions. This temperature change could impact the ratio of ice to water droplets within a mixed-phase cloud by delaying the onset of glaciation and by increasing the supercooled liquid fraction of the cloud, thereby modifying the radiative properties and the lifetime of the cloud. How are these aerosol-cloud interactions being impacted by photochemistry? Concurrently, a photomineralization mechanism was quantified by monitoring the loss of organic carbon and the simultaneous production of organic acids, such as formic, acetic, oxalic and pyruvic acids, CO and CO2. In more recent work, the role of indirect photochemistry including the in situ production of reactive oxygen species such as OH radicals, peroxides and singlet oxygen is being linked to the photomineralization mechanism. This mechanism explains and predicts the observed increase in CCN and decrease in IN efficiencies. Indeed, we have shown that photochemical processing can be a dominant atmospheric ageing process, impacting CCN and IN efficiencies and concentrations. Photomineralization can thus alter the aerosol-cloud radiative effects of organic matter by modifying the supercooledliquid-water-to-ice-crystal ratio in mixed-phase clouds with implications for cloud lifetime, precipitation patterns and the hydrological cycle.



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