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## Abstract

## Phase Transitions in Atmospheric Aerosols and their Importance for Climate and Air Chemistry

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Without phase transitions of condensable gases to the liquid or solid state the character of the Earth's atmosphere would be entirely different and its current supporting function for life on this planet would not exist. Besides water vapor itself there is a variety of condensable gases, such as sulfuric acid, nitric acid, ammonia, and a multitude of semivolatile organic species, which play a crucial role in atmospheric phase transitions. These gases determine the pathways for nucleation of aqueous aerosol droplets, also called gas-to-particle conversion, because they significantly lower the nucleation barrier. The resulting aerosol droplets serve as condensation nuclei, enabling the formation of liquid water clouds in the lower few kilometers of the atmosphere. At higher altitudes and lower temperatures the phase transition from water or aqueous solution droplets to ice crystals enables the formation of freezing of cloud droplets, accompanied by the release of great quantities of latent heat, profoundly influencing the motion of cloudy air masses and causing precipitation.

The modern understanding of these cloud formation and precipitation processes is centered on physico-chemical investigations of the homogeneous and heterogeneous nucleation processes, i.e. phase transitions without and with preexisting nucleus, respectively. The classical example of a homogeneous nucleation process is the bimolecular gas-to-particle conversion of H<sub>2</sub>SO<sub>4</sub> and H<sub>2</sub>O, which still today challenges experimentalists as well as theoreticians. Amongst the heterogeneous nucleation processes the importance of mineral dust particles as potent ice nuclei has been long recognized, but guantification of available amounts of these nuclei in the atmosphere and of their nucleation potential remains subject of Recently phase transitions in highly concentrated, almost water-free current debate. aerosols, have been shown to affect their ice nucleus capabilities, e.g. efflorescence and deliguescence representing transitions to or from water-free salt crystals or solid organics. In aerosols dominated in composition by organics another type of transition is that of liquid-liquid phase separation, which can occur as bimodal (nucleation and growth) or spinodal (barrierfree) phase transition. Micelle formation can occur in the presence of organic surfactants, as they exist for example in sea spray aerosols. Finally, at certain temperatures and relative humidities, organics-dominated aerosols can vitrify (i.e. enter a glassy state) of extreme viscosity and with massively suppressed molecular diffusivity in the glassy phase. The formation of a glassy state makes the aerosols inaccessible to many processes, such as water uptake, ice formation, and heterogeneous chemical reactions.