

THESIS FOR DEGREE OF DOCTOR OF PHILOSOPHY

Experimental studies of  
ice particle Formation Studies  
with Tropospheric Relevance

Erik Svensson

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University of Gothenburg and Chalmers University of Technology

Faculty opponent is Professor John Wettlaufer  
Nordic Institute for Theoretical Physics  
Stockholm, Sweden



UNIVERSITY OF GOTHENBURG

Department of Chemistry, Atmospheric Science  
University of Gothenburg  
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## Abstract

In their fourth assessment report, the Intergovernmental Panel on Climate Change points out the influence of particles suspended in air, known as an aerosol, as the main uncertainty in the understanding of the climate system. Aerosols affect climate in a number of ways. The so-called direct effect is due to scattering and absorption of light passing through the atmosphere. The indirect effects are caused by aerosol-cloud interactions. The formation of clouds requires pre-existing particles for the water vapor to condense onto. The number concentration, size distribution and chemical properties, as well as the atmospheric dynamics, determine the development of a cloud. Clouds reflect incoming solar radiation into space, but also trap heat radiation from the ground. Ice formation in clouds affects their interaction with radiation, their lifetime, and it may induce the formation of precipitation. Cloud droplets can be supercooled to very low temperatures, and droplets of pure water freeze by homogeneous nucleation at temperatures below  $-33^{\circ}\text{C}$ . However, droplet freezing can be facilitated by solid particles, a process called heterogeneous freezing.

This thesis presents results from laboratory studies of ice formation processes of atmospheric relevance. The overall aim has been to improve the understanding of ice cloud microphysics. An electrodynamic balance was developed and used to study freezing of single levitated droplets in the micrometer size range. Evaporation freezing of oxalic acid solution droplets was investigated at temperatures from 236 to 248 K. Freezing was observed in the whole temperature range, but at the higher temperatures freezing took place after a period of droplet evaporation. The process was explained by the formation of oxalic acid precipitates as the droplets evaporated, which in a subsequent step induced freezing. The potential importance of the process in atmosphere is currently unknown, but it may explain observed freezing in evaporating cloud and further studies should elucidate if the process is a general phenomenon for a large group of compounds. The same setup was also used to study freezing induced by kaolinite particle in collisions with pure water droplets. One or a few collisions were usually sufficient to induce freezing at temperatures from 240 to 268 K, and relative humidity was observed to be important for the freezing efficiency.

Aerosol and cloud studies were carried out with a bistatic lidar at the ALOMAR research facility in Andenes, Norway. The degree of linear polarization calculated from the experiments was compared with different types of light scattering calculations. Aerosol results obtained below 3.6 km could only be explained by non-spherical particles. The results obtained for thin clouds could be simulated by ice particles with a certain degree of surface roughness. The bistatic lidar method may provide a useful technique for the development of an improved cirrus cloud climatology and it may be used to validate other techniques.

Additional laboratory experiments were carried out to study ice formation on graphite, which was used as a proxy for soot particles in the atmosphere. The presence of dinitrogen pentoxide and nitric acid was concluded to facilitate the growth of thick ice layers and the process was sensitive to temperature and gas deposition rate. The experiments provide an improved molecular level understanding of ice formation, and a starting point for further research on systems of increased complexity.