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Infrared spectra of nitric acid hydrates - influence of particle shape

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Lidar observations as well as mid infrared remote sensing instruments have become valuable tools to analyse composition and phase of polar stratospheric cloud (PSC) constituents. At temperatures above the ice frost point, a strong depolarization in the lidar measurements clearly indicates the presence of aspherical nitric acid hydrate crystals, either nitric acid dihydrate (NAD) or nitric acid trihydrate (NAT), as opposed to spherical supercooled ternary solution droplets (STS) of water, sulphuric acid, and nitric acid. Nonetheless, infrared spectra of nitric acid hydrates are still commonly analysed, both in laboratory and in field studies, applying Mie theory for spheres. In this presentation, we will show that the extinction spectra of nitric acid hydrates are subject to strong asphericity-induced spectral changes so that approximating their particle shape by spheres might lead to erroneous retrieval results.

Our study is based on recent simulation experiments at the AIDA aerosol and cloud chamber of Forschungszentrum Karlsruhe (Aerosol Interaction and Dynamics in the Atmosphere), focussing on different pathways for the formation of NAD particles at temperatures around 195 K. First of all, we generated NAD particles or, more precisely, alpha-NAD crystals by shock freezing of a nitric acid/water gas mixture. In this case, Mie theory proved to be adequate to accurately reproduce the concomitantly recorded infrared spectrum of the airborne alpha-NAD particles. On the contrast, Mie theory clearly failed to reproduce the extinction features of alpha-NAD crystals obtained via homogeneous nucleation of supercooled nitric acid solution droplets, freezing and growing slowly on a time scale of several hours. Our measured infrared spectrum could only be accurately fitted with the help of T-matrix calculations assuming

oblate particles with aspect ratios greater than five. Since homogeneous nucleation in supercooled STS droplets is one suggested pathway for the formation of solid nitric acid hydrates in the winter polar stratosphere, also strongly aspherical particle shapes should be considered when analysing lidar or infrared spectral data.

As we never observed any spectral indication of NAT formation in our nucleation experiments (at NAT saturation ratios of up to 26 and on time scales of up to four hours), our spectroscopic study is limited to nitric acid dihydrate crystals. However, we will also present T-matrix calculations of the infrared spectrum of NAT to analyse the asphericity-induced spectral changes in the infrared extinction features of this hydrate.