Probing ice clouds by mid-infrared extinction spectroscopy: case studies from ice nucleation experiments in the AIDA aerosol and cloud chamber

Series of infrared extinction spectra of ice crystals were recorded in the 6000–800 cm−1 wavenumber regime during expansion cooling experiments in the large aerosol and cloud chamber AIDA of Forschungszentrum Karlsruhe. Either supercooled sulphuric acid solution droplets or dry mineral dust particles were added as seed aerosols to initiate ice formation after having established ice supersaturated conditions inside the chamber. The various ice nucleation runs were conducted at temperatures between 237 and 195 K, leading to median sizes of the nucleated ice particles of 1–15 μm. The measured infrared spectra were fitted with reference spectra from **T**-matrix calculations to retrieve the number concentration as well as the number size distribution of the generated ice clouds. The precise evaluation of the time-dependent ice particle number concentrations, i.e., the rates of new ice particle formation, is of particular importance to quantitatively analyse the ice nucleation experiments in terms of nucleation rates and ice activation spectra. The ice particles were modelled as finite circular cylinders with aspect ratios ranging from 0.5 to 3.0. Benefiting from the comprehensive diagnostic tools for the characterisation of ice clouds which are available at the AIDA facility, the infrared retrieval results with regard to the ice particle number concentration could be compared to independent measurements with various optical particle counters. This provided a unique chance to quantitatively assess potential errors or solution ambiguities in the retrieval procedure which mainly originate from the difficulty to find an appropriate shape representation for the aspherical particle habits of the ice crystals. Based on these inter-comparisons, we demonstrate that there is no standard retrieval approach which can be routinely applied to all different experimental scenarios. In particular, the concept to account for the asphericity of the ice crystals, the a priori constraints which might be imposed on the unknown number size distribution of the ice crystals (like employing an analytical distribution function), and the wavenumber range which is included in the fitting algorithm should be carefully adjusted to each single retrieval problem.
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