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A novel model to predict the physical state of atmospheric $H_2SO_4/NH_3/H_2O$ aerosol particles

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Abstract. The physical state of the tropospheric aerosol is largely unknown despite its importance for cloud formation and for the aerosol's radiative properties. Here we use detailed microphysical laboratory measurements to perform a systematic global modelling study of the physical state of the $H_2SO_4/NH_3/H_2O$ aerosol, which constitutes an important class of aerosols in the free troposphere. The Aerosol Physical State Model (APSM) developed here is based on Lagrangian trajectories computed from ECMWF (European Centre for Medium Range Weather Forecasts) analyses, taking full account of the deliquescence/efflorescence hysteresis. As input APSM requires three data sets: (i) deliquescence and efflorescence relative humidities from laboratory measurements, (ii) ammonia-to-sulfate ratios (ASR) calculated by a global circulation model, and (iii) relative humidities determined from the ECMWF analyses. APSM results indicate that globally averaged a significant fraction (17-57%) of the ammoniated sulfate aerosol particles contain solids with the ratio of solid-containing to purely liquid particles increasing with altitude (between 2 and 10 km). In our calculations the most abundant solid is letovicite, $(NH_4)_3H(SO_4)_2$, while there is only little ammonium sulfate, $(NH_4)_2SO_4$. Since ammonium bisulfate, NH_4HSO_4, does not nucleate homogeneously, it can only form via heterogeneous crystallization. As the ammonia-to-sulfate ratios of the atmospheric $H_2SO_4/NH_3/H_2O$ aerosol usually do not correspond to the stoichiometries of known crystalline substances, all solids are expected to occur in mixed-phase aerosol particles. This work highlights the potential importance of letovicite, whose role as cloud condensation nucleus (CCN) and as scatterer of solar radiation remains to be scrutinized.

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