Evaporation and condensation of semivolatile aerosol compounds in the DMT-CCN counter

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Atmospheric aerosol particles are composed of large variety of chemical compounds with different vapour pressures affecting the partitioning between aerosol and gas phase. Some of these can be considered to be nonvolatile meaning their vapor pressure is so low that in practice they are always found from the particle phase. Other group of compounds found from particles is semivolatile. Depending on the conditions, like temperature and relative humidity, these compounds may exist in the gas, liquid or solid particle phase. This has to be taken into account both to fully address the effect of semivolatiles on indirect and indirect aerosol forcing, and also in the experiments when hygroscopicity and CCN activity of aerosol particles are addressed.

In this study the effect of inorganic semivolatile aerosol compounds on CCN activity of aerosol particles is studied by using computational model for DMT-CCN counter (Roberts and Nenes 2005), cloud parcel model for condensation kinetics and experiments to quantify the modeled results. Concentrations of water vapour and semivolatiles as well as aerosol trajectories in the CCNcolumn are calculated by a computational fluid dynamics model (Raatikainen et al. 2012). These trajectories and vapor concentrations were then used as an input for cloud parcel model (Romakkaniemi et al. 2005) to simulate condensation kinetics of water and semivolatiles between aerosol particles and the gas phase. From these results the smallest particle size able to activate in different supersaturations was determined. This size is called as a critical particle diameter. From measurements this is determined as a particle size where 50% of particles activated in the CCN counter.

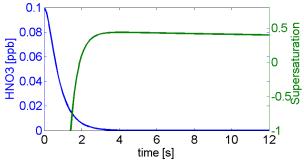


Figure 1: Nitric acid concentration and water supersaturation in the CCN column as a function of time in the column.

In the experimental and modeling setup the dried ammonium nitrate particles are taken into CCN column in the CCN counter. Beyond being the source for water, wetted walls in the CCN column act as a perfect sink for gas phase ammonia and nitric acid. Thus their gas phase concentration (Figure 1) is quickly decreased in the centerline of sample flow in the direction of flow. Depending on the gas phase concentration of nitric acid and ammonia, they start condense on particles or evaporate from particles as soon as the aerosol will become liquid solution.

Table 1. Comparison between modelling and experimental results for the critical aerosol particle diameters in different supersaturations.

Supersa	Meas	Mod	Mod Dc [nm]
turation	Dc [nm]	Dc [nm]	no evap.
0.2	84	91	80
0.3	71	75	62
0.4	62	65	51
0.6	53	54	40
0.8	47	48	33

As can be seen from the results presented in Table 1, the modelled and measured critical diameters agree nicely when evaporation is taken into account. Thus the model is able to accurately predict the evaporation of nitric and ammonia from aerosol particles before the maximum supersaturation is reached. Based on these results evaporation of semivolatile aerosol compounds can lead to underestimation of aerosol CCN activity.

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