

Uptake of N₂O₅ to citric acid aerosol particles

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Dinitrogen pentoxide is a significant reactive intermediate in the night time chemistry of nitrogen oxides, serving as a major NO_x sink by heterogeneous hydrolysis on aerosol surfaces. It can influence tropospheric ozone production and therefore the oxidative capacity of the atmosphere (Chang et al., 2011).

We used the short-lived radioactive tracer ¹³N (Ammann, 2001) in conjunction with an aerosol flow tube reactor in order to study N₂O₅ uptake kinetics on aerosol particles. ¹³NO was mixed with non labeled NO and O₃ in a first flow reactor, where N₂O₅ is synthesized under dry conditions. The N₂O₅ flow is then admitted into the aerosol flow reactor together with the aerosol flow containing deliquesced citric acid particles generated by ultrasonic nebulisation and conditioned to the relative humidity of interest. Movable inlets allow varying the length of the aerosol flow tube. Behind the reactor a Scanning Mobility Particle Sizer (SMPS) system is used to characterize the aerosol. The remaining gas exiting the reactor is passed through a parallel plate diffusion denuder system for selective separation of gaseous species and a particle filter to trap the particles. Activity of the ¹³N labeled species can be monitored via scintillation counters placed on the denuder plates and the particle filter, respectively.

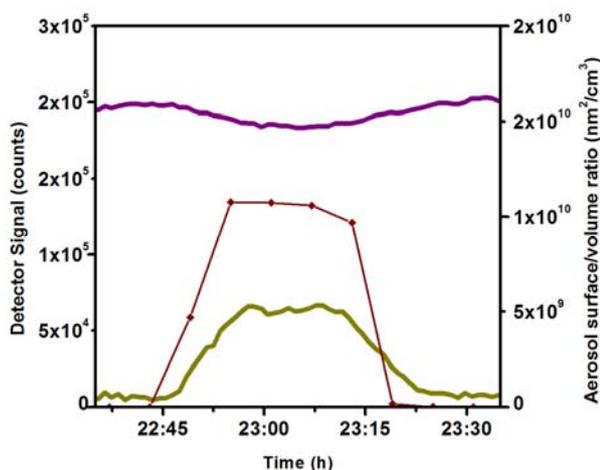


Figure 1. Gas phase N₂O₅ (solid magenta), particulate N (solid green, magnified by a factor of 10), and aerosol surface area (red) to volume ratio during an uptake experiment.

Fig. 1 shows data from a typical experiment with citric acid aerosol, where the loss of N₂O₅ from the gas phase is accompanied by an increase in particulate nitrate. Figure 2, shows a set of measurements conducted at a relative humidity of 62 %, at a fixed reaction time of

around one minute, and at varying aerosol surface to volume ratio, along with a simple first-order fit (Vlasenko et al., 2006).

The preliminary results obtained have shown that the uptake coefficient increases with humidity from $1.65 \pm 0.3 \times 10^{-3}$ (~27% RH) to $1.25 \pm 0.3 \times 10^{-2}$ (45% RH) and $2.00 \pm 0.3 \times 10^{-2}$ (61.5% RH). This is comparable to some polycarboxylic acids (like malonic acid), but higher than some others (Ammann et al., 2012). The increase is likely related to the increasing amount of water associated with citric acid at higher humidity that promotes hydrolysis of N₂O₅.

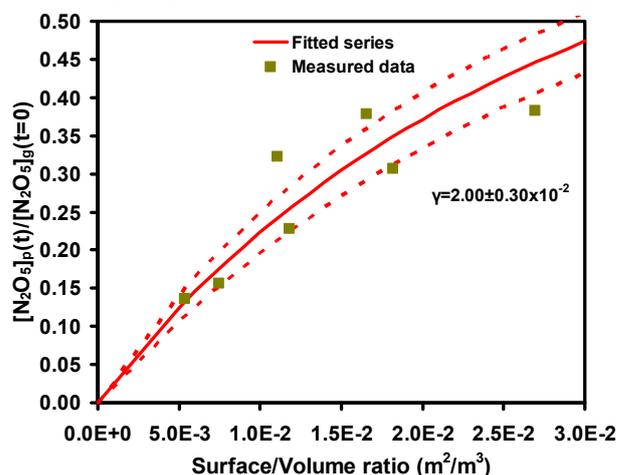


Figure 2. Data set collected using citric acid aerosol at 61.5% RH. Green squares represent measured data points while the red curve is a fit to the data according to the kinetic model, with the dotted curve showing the error range

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