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# Temperature dependence of heterogeneous uptake of N<sub>2</sub>O<sub>5</sub> by ammonium sulfate aerosol

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## Abstract

We present laboratory measurements of the dependence of the uptake coefficient  $\gamma$  on temperature for removal of the NO<sub>x</sub> reservoir N<sub>2</sub>O<sub>5</sub> by sub-micron aerosols containing ammonium sulfate and ammonium hydrogen sulfate. Uptake coefficients were measured over the range 263–303 K at 50% RH, which varied in the range 0.005–0.036. A simple negative dependence of uptake on temperature is observed for ammonium sulfate over this range, while the form of the temperature dependence of uptake by ammonium hydrogen sulfate is more complex. The results consolidate those of an earlier less definitive study of the temperature effect on N<sub>2</sub>O<sub>5</sub> uptake. Copyright © 2009 Royal Meteorological Society

**Keywords:** N<sub>2</sub>O<sub>5</sub>; aerosol; temperature dependence; kinetics; sulfate

## 1. Introduction

The heterogeneous conversion of the NO<sub>x</sub> reservoir N<sub>2</sub>O<sub>5</sub> to HNO<sub>3</sub> on aerosol surfaces is an important loss process for nitrogen oxides at night. This has an effect on the oxidising capacity of the atmosphere as NO and NO<sub>2</sub> are involved in the catalytic processes that create ozone and hydroxyl. Several studies have investigated the importance of this process in the global troposphere (Dentener and Crutzen, 1993; Tie *et al.*, 2003; Evans and Jacob, 2005). All studies show a significant decrease in global NO<sub>x</sub> and ozone due to removal of N<sub>2</sub>O<sub>5</sub> onto aerosol particles. A critical parameter is the uptake coefficient (i.e. the fraction of collisions of N<sub>2</sub>O<sub>5</sub> with aerosol surfaces that results in irreversible loss). Laboratory measurements show that the uptake coefficient on aerosols typical of the troposphere depends in a complex manner with temperature, humidity and composition (Hu and Abbatt, 1997; Wahner *et al.*, 1998; Hallquist *et al.*, 2000; Thornton *et al.*, 2003). Evans and Jacob used these data to derive parameterisations of uptake as a function of relative humidity (RH) and temperature for use in the GEOS-CHEM model. This improved the agreement between models and measurements. The importance of heterogeneous removal was demonstrated by field measurements made during the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) campaign on the NOAA-P3 aircraft (Brown *et al.*, 2006), which showed a highly variable efficiency in the removal of N<sub>2</sub>O<sub>5</sub> by aerosol particles, depending on aerosol composition, as expected from laboratory data. Very recently, Bertram *et al.* have reported laboratory uptake measurements using ambient aerosol which indicate efficient removal, dependent on relative humidity and particle composition (Bertram *et al.*, 2008, personal communication).

The effect of temperature on the heterogeneous reaction arises directly from the effect on the uptake coefficient and indirectly on the relative humidity and hence on the water content of the aerosol. The study of Hallquist *et al.* (2003) remains the only published study to date of the dependence of uptake coefficient of N<sub>2</sub>O<sub>5</sub> on temperature, for conditions relevant to the lower troposphere. In that study, quite large experimental uncertainty was encountered, partly due to the difference between conditions of measurement of uptake rate, which was determined at the temperature of interest, and measurement of aerosol size distribution, which was performed at room temperature. In this work, we have employed a temperature-controlled differential mobility analyser, held at the same temperature and humidity as the aerosol flow tube, giving accurate aerosol size distributions under the conditions of the kinetics measurement. We report measurements of the temperature dependence of  $\gamma$  over the range 268–303 K, for ammonium sulfate and ammonium bisulfate aerosol.

## 2. Experimental

The experimental method is similar to that in previous work, but using a new apparatus for preparation and characterisation of the aerosol (Hallquist *et al.*, 2003; Badger *et al.*, 2006). Measurements were made using a flow tube apparatus over a range of temperatures. The kinetics apparatus is described in detail elsewhere (Badger *et al.*, 2006) and only the modifications are presented here.

The aerosol was generated at room temperature using a TSI 3076 atomiser containing bulk solutions of 0.1 M concentration of ammonium sulfate or hydrogen sulfate (bisulfate). A flow of approx. 1500 sccm

was used to generate sub-micron aerosol with aerosol number density in the range  $10^5$ – $10^7$  cm $^{-3}$ .

The aerosol was conditioned to the required humidity using a new custom-made unit (THC-A, Tapcon). The temperature and humidity of the aerosol were regulated by Peltier heat pump-controlled stages. The conditioning was performed prior to admission of the aerosol to the temperature-controlled region (inside a commercial freezer box) so that the humidity of the aerosol remained less than 100%. As far as is practical, the humidity was controlled by cooling the aerosol while keeping relative humidity within the desired range. This avoids the need to dilute the aerosol and allows control of the phase of deliquescent/efflorescent aerosol; although, at the lower end of the temperature range, where the water content of the aerosol is low, an additional dry flow of nitrogen was required to be added to the aerosol stream to achieve the required humidity.

After conditioning, the aerosol flow entered the freezer box in which both the differential mobility analyser (DMA) and kinetics apparatus were held. Here the flow was divided: a portion of the flow (1000 sccm) was passed to the DMA for characterisation, while around 500 sccm was sent to the aerosol flow tube. A plenum volume, open to atmospheric pressure, allowed excess flow to escape to a vent. The aerosol size distribution was characterised using a Tapcon DMA, coupled to a Faraday cup electrometer (FCE-08A). The design and performance of the DMA is described elsewhere (Winklmayr *et al.*, 1991). The DMA was operated with sheath air at the same humidity as the input aerosol stream. A separate THC unit operated in recirculation mode controlled the humidity of the sheath air. The size distribution was therefore determined at the same temperature and humidity as the kinetics measurements, which avoids errors introduced in previous work arising from evaporation of aerosol water (Kane *et al.*, 2001) or from the need to correct the aerosol surface area, measured at room temperature, for decreased temperature and increased humidity (Hallquist *et al.*, 2003).

The concentration of N<sub>2</sub>O<sub>5</sub> within the flow tube was determined using the chemiluminescence technique described in Hallquist *et al.* (2000), wherein the N<sub>2</sub>O<sub>5</sub> is quantitatively thermally decomposed to NO<sub>3</sub> and the nitrate radical then titrated by NO to NO<sub>2</sub>. The concentration of N<sub>2</sub>O<sub>5</sub> is thereby derived as the change in NO concentration on admission of N<sub>2</sub>O<sub>5</sub> to the flow tube.

Kinetics measurements were performed in a laminar-flow aerosol flow tube by measuring first-order loss rate coefficient of N<sub>2</sub>O<sub>5</sub> in the presence and absence of aerosol. As in previous work, the internal surfaces of the flow tube were coated with halocarbon wax, with the result that the uptake to the walls was reduced by an order of magnitude. Uptake coefficients  $\gamma_w$  for removal onto the walls were of the order of  $2 \times 10^{-6}$ , similar to those used in previous work (Griffiths *et al.*, 2008).

The contact time between the N<sub>2</sub>O<sub>5</sub> and the aerosol was controlled remotely using a new custom-built sliding injector (Tapcon, SI-10). A linear actuator controlled the sliding injector and its position was measured using a calibrated sensor. This allowed for precise, reproducible control of contact time to better than 0.1 s. For these measurements, a small ‘pepper-pot’ was placed over the end of the sliding injector to promote mixing.

The uptake coefficient  $\gamma$  derived from the rate of loss of N<sub>2</sub>O<sub>5</sub> within the flow tube is, using Equation (1):

$$-\frac{d[N_2O_5]}{dt} = k^I [N_2O_5] = \gamma \cdot S_a \cdot \frac{c}{4} [N_2O_5] \quad (1)$$

where  $S_a$  is the surface area density (derived from the DMA measurements),  $c$  the average molecular speed of N<sub>2</sub>O<sub>5</sub> in the gas phase and  $[N_2O_5]$  the concentration of N<sub>2</sub>O<sub>5</sub>. Mixing ratios within the flow tube were between 100 and 400 ppbv. The first-order rate coefficient was derived using the measured loss rate coefficients in the presence and absence of aerosol according to the procedures described by Brown (1978).

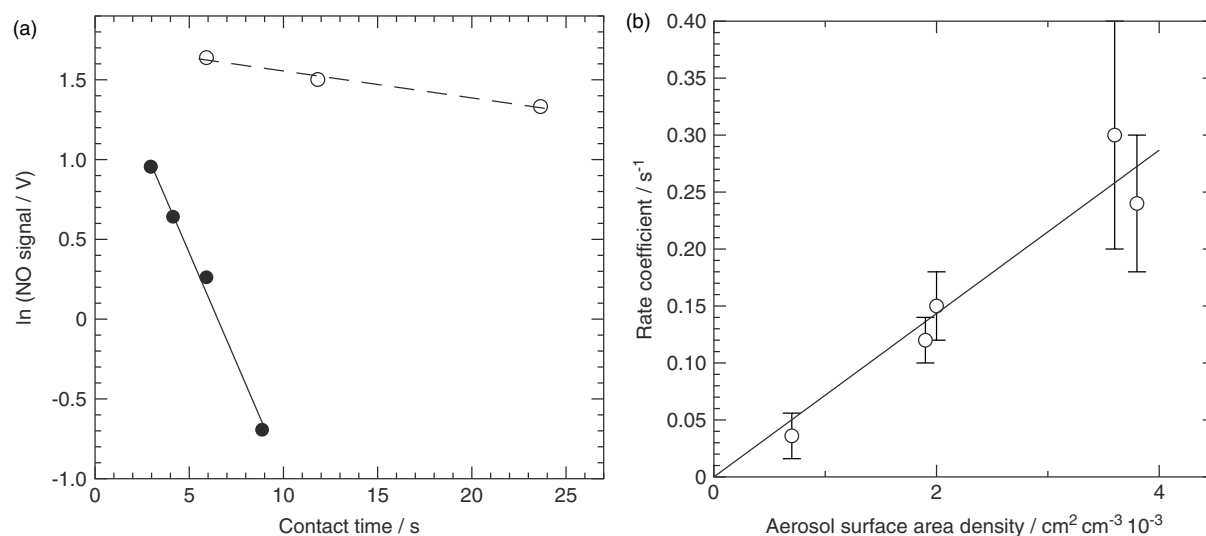
Under our experimental conditions of a linear flow velocity of around 0.7 cm s $^{-1}$ , the Reynold’s number was 16 and the diffusional mixing time was 3 s. First-order decays were observed independent of the time over which the data were measured over the range 3–20 s, indicating efficient mixing, presumably due to the effects of the turbulent flow produced at the exit of the ‘pepper pot’.

Experiments were performed over a range of N<sub>2</sub>O<sub>5</sub> mixing ratios and aerosol surface areas. Typically, N<sub>2</sub>O<sub>5</sub> mixing ratios were around 300 ppbv and aerosol surface area density varied in the range  $1 \times 10^{-4}$ – $5 \times 10^{-3}$  cm $^2$  cm $^{-3}$ . The aerosol volume to surface ratio varied between  $3 \times 10^{-9}$  and  $4 \times 10^{-9}$  m.

Figure 1(a) shows the experimental data gathered during a measurement uptake of N<sub>2</sub>O<sub>5</sub> by ammonium sulfate at 293 K. The data are plotted in log-linear form. Figure 1(b) shows a plot of  $k^I$  versus aerosol surface area density, and is consistent with low systematic errors in the measurements. Uptake was measured as a function of temperature over the range 263–303 K, and relative humidity was 50% throughout the measurements.

### 3. Results

Table I summarizes the uptake coefficients for ammonium sulfate and ammonium hydrogen sulfate over the range 263–303 K, measured in this work. The uncertainty in the derived uptake coefficient is dominated by uncertainty in the measurement of N<sub>2</sub>O<sub>5</sub> mixing ratio within the flow tube, which was 10–20 ppbv. Depending on the conditions used, the uptake rate and the aerosol surface area, this introduces an uncertainty of at most 25% for a single determination of  $\gamma$ .



**Figure 1.** (a) Kinetics of uptake of N<sub>2</sub>O<sub>5</sub> by ammonium sulfate aerosol at 293 K and 50% RH – open symbols/broken line: loss to reactor walls; filled symbols/solid line: loss to aerosol. (b) Dependence of measured uptake rate coefficient on aerosol surface area density. Data shown for ammonium sulfate aerosol at 293 K and 50% RH.

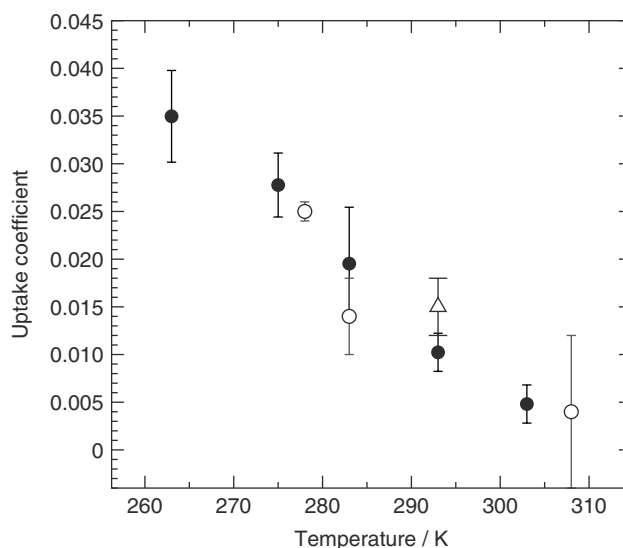
**Table I.** Uptake coefficients for heterogeneous uptake of N<sub>2</sub>O<sub>5</sub> by ammonium sulfate and bisulfate aerosol at 50% RH.

Ammonium sulfate		
Temperature (K)	Uptake coefficient	Number of determinations
263	0.035 ± 0.002	2
275	0.028 ± 0.002	4
283	0.020 ± 0.005	2
293	0.010 ± 0.002	3
303	0.005 ± 0.002	2
Ammonium bisulfate		
263	0.036 ± 0.009	3
268	0.005 ± 0.002	2
278	0.014 ± 0.006	5
283	0.012 ± 0.001	3
288	0.011 ± 0.001	2
293	0.016 ± 0.003	2
303	0.003 ± 0.001	2

The uptake coefficients reported in Table I are calculated from the mean of the individual determinations, weighted by the uncertainty of the individual uptake coefficients. The error quoted is  $2\sigma$ , where  $\sigma$  is the standard error of the mean. The measurements from this work are plotted in Figures 2 and 3 together with those reported previously by Hallquist *et al.* (2003) and Griffiths *et al.* (2009).

#### 4. Discussion

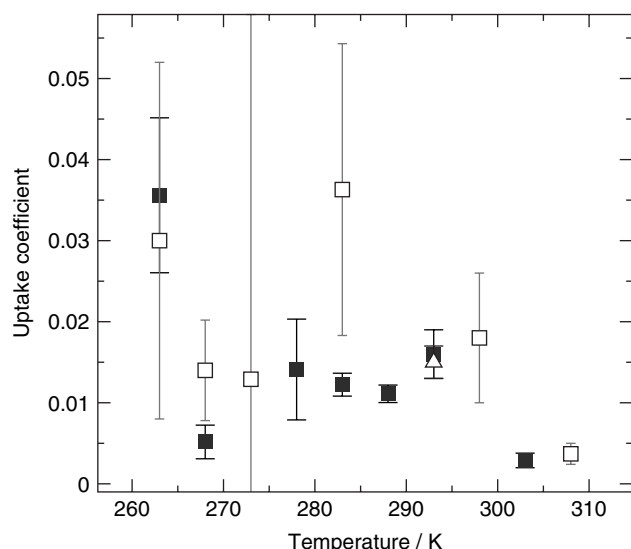
The uptake coefficients at 50% RH are in good agreement with the previous determinations (Hallquist *et al.*, 2003), with improved precision reducing the uncertainty in the observed temperature trend. The quality of the data has been improved by performing aerosol uptake and aerosol characterization under the



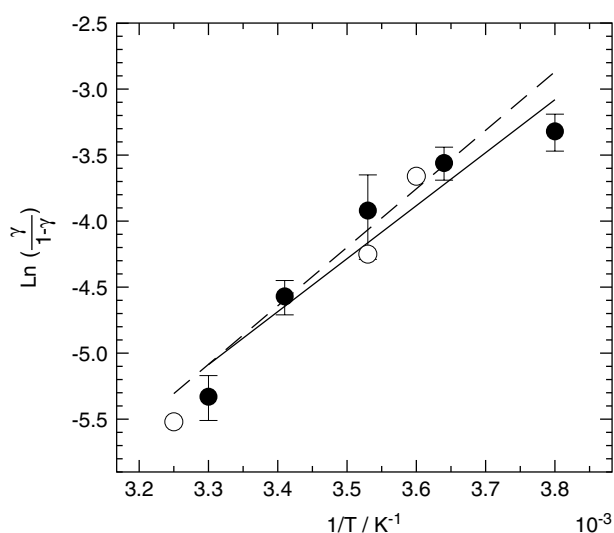
**Figure 2.** Uptake coefficients for heterogeneous uptake of N<sub>2</sub>O<sub>5</sub> by ammonium sulfate aerosol. Filled circles: this work; open circles: Hallquist *et al.* (2003); triangle: Griffiths *et al.* (2008).

same conditions of temperature and humidity. Additionally, halocarbon wax coating reduced the rate of loss to the reactor walls and allowed measurement of small aerosol uptake rate coefficients, while the computer-controlled sliding injector allowed for precise control of contact times.

The results confirm that heterogeneous uptake of N<sub>2</sub>O<sub>5</sub> by sulfate aerosol is a strong function of temperature over the range 263–303 K. There is a general negative dependence of the uptake coefficient on temperature, which is larger than that for uptake into sulfuric acid H<sub>2</sub>SO<sub>4</sub> aerosol (Hallquist, 2000). Also, the behaviour of the two ammonium salts is qualitatively different over the range 263–303 K, with ammonium sulfate showing a monotonically increasing uptake coefficient with decreasing temperature,



**Figure 3.** Uptake coefficients for heterogeneous uptake of  $\text{N}_2\text{O}_5$  by ammonium bisulfate aerosol. Filled squares: this work; open squares: Hallquist *et al.* (2003); triangle: Griffiths *et al.* (2008).



**Figure 4.** Plot of  $\ln(\gamma/(1-\gamma))$  versus  $1/T$  for uptake on ammonium sulfate. The gradient of this plot gives  $\Delta H/R$  and the intercept  $\Delta S/R$ . Data are shown for uptake by ammonium sulfate. Solid symbols: this work; open symbols: Hallquist *et al.*, 2003. Solid line: fit of Equation (2) to the present data; broken line: fit of all data to Equation (2).

while the behaviour of the ammonium bisulfate system is more complex. The results consolidate these unexpected differences, which were apparent in an earlier, less definitive study of the temperature effect on  $\text{N}_2\text{O}_5$  uptake (Hallquist *et al.*, 2003).

In general, uptake is the result of a series of coupled processes, including gas-to-particle transfer (accommodation), incorporation into the bulk and reaction with the constituents of the aerosol. Within the resistor model, the uptake coefficient for a reactive process such as hydrolysis can be expressed as

$$\frac{1}{\gamma} = \frac{1}{\alpha} + \frac{1}{\Gamma_{\text{sol}} + \Gamma_{\text{rxn}}} \quad (2)$$

In the above, the first term represents the resistance due to gas–particle transfer so that  $\alpha$  is the mass accommodation coefficient representing the probability of entry of the gas molecule into the aerosol particle. The second term represents the coupled reaction–diffusion processes wherein,  $\Gamma_{\text{sol}}$  the resistance due to limited solubility of the reactant gas, and  $\Gamma_{\text{rxn}}$  the resistance due to reaction within the aerosol volume. When reaction within the aerosol is fast, accommodation at the surface becomes rate limiting and  $\gamma \cong \alpha$ .

The temperature dependence of the uptake coefficient can be used to identify which of the coupled process is rate-determining. A thermodynamic model of mass accommodation has been developed by Davidovits and co-workers, in which the accommodation coefficient is related to the free energy change on accommodation according to (Davidovits *et al.*, 1995).

$$\frac{\alpha}{1-\alpha} = \exp\left(\frac{-\Delta G^*}{RT}\right) \quad (3)$$

The temperature dependence of the reactive uptake coefficients of  $\text{N}_2\text{O}_5$  on ammonium sulfate aerosol is consistent with accommodation-controlled uptake. An analysis of the data for ammonium sulfate, using Equation (3) and assuming that  $\gamma = \alpha$ , yields values of  $\Delta H_{\text{obs}} = -33 \text{ kJ/mol}$  and  $\Delta S_{\text{obs}} = -152 \text{ J K}^{-1} \text{ mol}^{-1}$  for the enthalpy and entropy of accommodation for ammonium sulfate. Figure 4 shows a fit of the data to Equation (3). These values are larger than those obtained for the temperature dependence of uptake of on sulfuric acid aerosol ( $\Delta H_{\text{obs}} = -25 \text{ kJ mol}^{-1}$  and  $\Delta S_{\text{obs}} = -115 \text{ J K}^{-1} \text{ mol}^{-1}$ , Hallquist *et al.*, 2000), but similar to those derived from the temperature dependence of uptake on pure water droplets of  $\Delta H_{\text{obs}} = -33.5 \text{ kJ mol}^{-1}$  and  $\Delta S_{\text{obs}} = -156 \text{ J K}^{-1} \text{ mol}^{-1}$  (George *et al.*, 1994). Both these uptakes are believed to be accommodation-controlled but hydrolysis in acid solution occurs by an acid-catalysed mechanism, which would not be favoured in ammonium sulfate.

The picture emerging from the data for ammonium bisulfate at 50% RH is less clear. Over the range of temperatures studied, a simple monotonic behaviour, as predicted by Equation (3), is not observed. The complex dependence on temperature is outside the experimental uncertainty and is not clearly identifiable with any single process. It is clear from the data that the resistance due to solvation/reaction and diffusion becomes significant at  $T < 290 \text{ K}$  for this system. While calculations using the Aerosol Inorganic Model (<http://www.hpc1.uea.ac.uk/~e770/aim.html>) indicate that the formation of solid letovicite  $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$  in the aerosol particles is possible below 290 K, the aerosol mass fraction of letovicite is not correlated with the uptake coefficient over the range 290–263 K. However, the much larger uptake coefficient at the lowest temperature (263 K) could be due to kinetic

restriction on the phase change to letovicite, leading to supersaturated aqueous particles at  $T > 290$  K.

## 5. Atmospheric significance

From these measurements, it is clear that the temperature dependence of the uptake coefficient is significant for lower tropospheric conditions. Recent field studies and ambient aerosol studies indicate suppression of the uptake rate under ambient conditions due to low RH (crystallisation), the presence of organics and possibly the nitrate effect (Bertram *et al.*, 2009), all of which are consistent with laboratory data. The increase in uptake coefficient with decrease in temperature would compensate for these suppressing factors depending on the particular conditions prevailing and must be considered to quantitatively account for N<sub>2</sub>O<sub>5</sub> lifetime.

In the atmosphere, temperature and relative humidity vary strongly from the top to the bottom of the boundary layer. From our results, the uptake coefficient for ammonium sulfate increases from 0.01 to 0.02 on decreasing the temperature from 293 to 283 K. This is equivalent to transporting an air parcel from the ground to the top of the boundary layer and will, for an initial RH of 50% at 293 K, result in a final RH at 283 K of approximately 90%. Based on the hygroscopic growth factor of 1.5 of ammonium sulfate at 90% RH (Tang and Munkelwitz, 1993), the aerosol surface area will therefore increase on ascent by a factor of 3. Taken together, these two factors will increase the overall loss rate by a factor of 6, and this will have a marked impact on the heterogeneous loss of N<sub>2</sub>O<sub>5</sub>.

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