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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Toward a general parameterization of N_2O_5 reactivity on aqueous particles: the competing effects of particle liquid water, nitrate and chloride" *by* T. H. Bertram and J. A. Thornton

Anonymous Referee #1

Received and published: 12 August 2009

This manuscript presents new experiments on the heterogeneous reaction of N_2O_5 on mixed organic-inorganic aerosol particles using an established method, an aerosol flow tube coupled to a chemical ionization mass spectrometer. From these experiments the reactive uptake coefficient (γ) is determined in detail as a function of particle liquid water, nitrate, and chloride content. The measurements show that trace amounts of chloride can lead to N_2O_5 uptake in presence of high nitrate concentrations. The results are used to derive a new parameterization which accounts for the salting out effect by nitrate, the impact of chloride, and the amount of available particle water on the

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corresponding heterogeneous reaction kinetics. This parameterization is discussed with respect to previous descriptions of the heterogeneous N_2O_5 hydrolysis.

General comments:
The detailed analysis of the heterogeneous N_2O_5 hydrolysis and corresponding parameterization fit nicely within the scope of ACP and warrant publication. The manuscript is well structured and reads well. The experimental methods are sound. I recommend this manuscript for publication in ACP after the authors have addressed the minor comments given below.

Specific comments:

Introduction:
Page 15182, 1st paragraph: Although Dentener and Crutzen implemented first heterogeneous N_2O_5 hydrolysis for tropospheric conditions, recent studies such as Evans and Jacob should also be mentioned in the introduction and not only at the end of the manuscript. As pointed out at the end of the manuscript, Evans and Jacob implemented various particle types in their modeling study compared to previous work.
Page 15183, end of 1st paragraph: The work of others (e.g. Park et al. 2007, Anttila et al. 2006, Knopf et al. 2007, Cosman et al. 2008) which also indicate a strong variation of γ in presence of multicomponent aqueous solutions should be mentioned here.

Experimental design:
Page 15184, line 5: Studies by Park et al. 2007, Anttila et al. 2006, Knopf et al. 2007, Cosman et al. 2008 who determined a decrease in γ due to a coating should be mentioned as well.

Page 15185, line 18: You mean the resistivity of the water was greater than 18 MΩ cm?

Page 15186, line 1-3: The description of supplying either aerosol or particle free gas to the aerosol flow tube sounds a bit complicated, in particular “a two-state filter manifold system”. Maybe rewrite or simplify this section.

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Page 15187, line 21: Please give a reference to estimate the distance to establish a fully developed laminar flow.

Page 15188, line 1: Please give a reference for the diffusion constant for N_2O_5. How was it derived? Did you determine it experimentally?

Page 15188, line 4: The flow tube is 90 cm long. Why do you not use the last 50 cm of the tube?

Page 15189, line 18: Does the switching between filter bypass and filter inline has an effect on the measurement due to fluctuations in pressure and possibly RH or other factors?

Page 15191, line 15: Discussion Fig. 2b: I think that the statement of a "single unified description" may be too strong when looking at Fig. 2a and taking the typical scatter and uncertainties of the data into account. More data in Fig. 2a would stronger corroborate this statement.

Page 15195, Eq. 3-6. State before the introduction of the equations that you assume steady-state for the derivation.

Page 15198, line 10: What do you mean with "..is smaller, though not necessarily statistically different..."? Your uncertainty is $\pm 1\sigma$? Your value is significantly smaller than a previous study. Elaborate on this.

Page 15207, Table 1: If the table contains values from a previous study (e. g. Thornton and Abbatt), please indicate these experiments as footnotes.

Technical comments:
When referring to the specific reactions use a full sentence. Often your reference follows at the end of the sentence. E.g. "....forming nitric acid (HNO_3) Reaction (R3)." Maybe insert "as indicated by Reaction (R3)".

In some locations a comma is missing in front of "and" or "or" when describing a series

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such as "H_2O, Cl^-, and/or NO_3^-".

Page 15184, R1–R4: The phase of the compounds should be given in normal font not as subscript.

Page 15190, line 5: Description of Eq. 2. Discard one time the expression "where".

Page 15203, line 1, 28 : there is a unnecessary line break.

Page 15204, lines 10, 28, 32 : there are unnecessary line breaks.

Figures:
Generally, the figure quality in the print version of this ACPD article was not satisfactorily. This may be due to the formatting or scaling of the images.

Fig. 1: The quality of the font and maybe its size should increase.

Fig. 2 - 5: The figures are too small in this print version and the font quality is not sufficient. The legends are hardly readable. I recommend using open and filled symbols to discriminate the different data sets.

Fig. 6: The legend "This study. . .." is not necessary.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15181, 2009.