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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

T. H. Bertram and J. A. Thornton

thbertram@ucsd.edu

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The interactive discussion of our paper, "Toward a general parameterization of N2O5 reactivity on aqueous particles: the competing effects of particle liquid water, nitrate and chloride" on ACPD raised a series of technical issues that we address in detail below. The final version of the manuscript, to be published in ACP, will reflect these changes. We thank the two anonymous reviewers for their helpful suggestions.

Response to comments from referee #1:

1. Page 15182, 1st paragraph: Although Dentener and Crutzen implemented the first heterogeneous N2O5 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.

References to Evans and Jacob, Riemer et al., and Davis et al. have been added to the introduction.

2. 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 gamma in the presence of multicomponent aqueous solutions should be mentioned here.

References to Park, Antilla, Knopf and Cosman have been added to the introduction as well.

3. 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 is due to a coating should be mentioned as well.

Again, references to Park, Antilla, Knopf and Cosman have been added to the discussion of the effect of coatings on gamma.

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

Yes, this has been added to the manuscript

5. 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.

While the statement is a bit complex, it is fully referenced and an accurate description of

the system. For clarity, we have modified the sentence to read ". . . or directed through a two-state filter manifold system designed to modulate the flow direction between filter-inline and filter-bypassed states for calculation of the N2O5 loss rate via the approach described in Bertram et al. [2009]." Further, we have reworded the text in the section comparing the two analysis techniques (section 2.3) for determining khet. 6. Page 15187, line 21: Please give a reference to estimate the distance to establish a fully developed laminar flow.

We have added a reference to Kay and Nedderman, Fluid Mechanics and Transfer Processes, Cambridge University Press, Cambridge, 1985.

7. Page 15188, line 1: Please give a reference for the diffusion constant for N2O5. How was it derived? Did you determine it experimentally?

The diffusion constant was taken from the estimate of Hu and Abbatt [1997], where they calculated it from standard Lennard-Jones potentials taken from Patrick et al. [1983] and Hirschfelder et al. [1964]. We have added reference to these here.

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

As stated in the manuscript, we use the central 50cm of the tube, i.e. 25 cm – 75 cm from the flow tube exit. This restriction is to ensure the reaction was probed under fully developed laminar flow and that the gas and particles were fully mixed.

9. 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?

Switching between the filter states does not alter the pressure in the flow reactor, as we use a filter substrate that has a very low pressure drop across the filter. The RH does change in some cases, often when the filter is heavily loaded with aerosol particles. However, in these cases we adjust the RH in the system so as to ensure that the RH

is the same in both filter states.

10. 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.

As shown in figure 2a, the measurements do not support a single RH dependence of gamma that is independent of particle composition. In contrast, as shown in figure 2b, we can describe gamma as a function of water molarity, independent of particle composition. We agree that the words used may be too strong. The statement now reads, "In contrast, when plotted as a function of particle water molarity, as determined by AIM, the dependence of gamma on water molarity is independent of particle chemical composition (Fig. 2b) to within the 1ïĄşïĂăexperimental uncertainty."

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

This has been done.

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

Agreed, to within the stated 1s uncertainty these values are statistically different. The text has been revised to read: "Our determination of k4/k3 is smaller than that inferred from the results of Behnke et al.(k4/k3=836±32) (1997)."

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

The values shown in the table are from this study only.

14. 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 (HNO3) 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 such as "H2O, Cl, and/or NO3 ".

We have searched the document and made these changes when appropriate.

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

We have made these changes.

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

We have made these changes.

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

We have made these changes.

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

We have made these changes.

19. 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.

We will address these issues with the ACP editorial staff. The figures provided were at least 300dpi, more than sufficient for submission. Nonetheless we will conform that the figures are readable in the final proof and make the necessary changes to ensure this. The legend in Figure 6 has been removed.
