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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Efficient bulk mass accommodation of N_2O_5 into neutral aqueous aerosol" *by* Goran Gržinić et al.

Anonymous Referee #2

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Review of Grzinic et al., Efficient bulk mass accommodation of N2O5 into neutral aqueous aerosol

Summary and General Comments: Grzinic et al presents a timely analysis of the reactive uptake of 13N labelled N2O5 to neutral aqueous nitrate and sulfate aerosol. The analysis permits assessment of N2O5 mass accommodation and the validity of the N2O5 disproportionation mechanism. The authors find high values for N2O5 mass accommodation (> 0.4) and the best evidence to date for the concerted N2O5 ionization mechanism, involving the nitronium ion intermediate. The results are a welcome addition to the field and should be published following the authors attention to the following general comments:

1) Most of the discussion centers on the uptake of N2O5 to nitrate aerosol. The con-

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Discussion paper

clusions drawn suggest large values for alpha and supports N2O5 ionization at the particle interface. What is less clear is if this is a general result for all aerosol at the same water activity. In the case of sulfate particles, labelled N2O5 likely evaporates from the interfacial region wo/the opportunity for exchange with unlabeled nitrate. The upper limit of 0.03 is achieved that is likely a combination of alpha, KH, and the reaction rates described. If KH and the hydrolysis rates are the same for sulfate and nitrate aerosol, is it correct to generalize the mass accommodation results derived from the nitrate aerosol experiments to all aqueous aerosol of comparable interfacial water activity? It would be helpful for the authors to provide some discussion on these points and the generality of the derived mass accommodation coefficient. I would find it very helpful if there was a second panel to Figure 3, which showed the processes for the sulfate particles.

2) Most of the aerosol flow reactor community is familiar with the kinetic equations shown, for unlabeled reactants. Are there any special considerations that need to be accounted for regarding single, vs multiple collisions of the labelled N2O5 with aerosol and the walls? Or rather, in Fig. 3, how many times would you expect a labelled N2O5 to cycle through a particle or the wall of the flow tube over the time constant of the flow reactor? Is this important to the analysis or derivation of the equations presented?

3) What is the effect of labelled HNO3 that is generated in the source region? How would this be interpreted in the experiment. It is not uncommon for N2O5 sources to be 10:1 HNO3 to N2O5. What is the expected ratio in this experiment?

4) Like reviewer #1, I am also confused by the notation of [N2O5]p. It would be helpful if equation 3 and Figure 2 were consistent in notation. It would also be helpful to denote between labelled and unlabeled here, as the unlabeled N2O5 uptake coefficient to nitrate aerosol could be pretty small even at high labeled N2O5 uptake coefficients, correct?

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1118, 2016.