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Discussions

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

Anonymous Referee #1

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This manuscript describes observations of the loss of radioactively labelled N2O5 upon exposure of this gas to neutral sulfate and nitrate aerosol. The hydrolysis reaction of N2O5 is one of the most important tropospheric heterogeneous reactions, leading to significant rates of NOx loss. Although the kinetics of this process have been studied for a long time there still remain interesting and important outstanding questions concerning its mechanism. This paper rightly addresses one of those issues, in particular the mass accommodation coefficient of N2O5 on aqueous particles. For context, in past studies it has been observed that reactive uptake coefficients tend to saturate at values somewhat larger than 0.01, suggesting that a mass accommodation coefficient of this size may be the limiting factor.

The authors use a unique experimental approach whereby they use short lived radioactively labelled N (N*) which is incorporated into N2O5, which is then injected into an

aerosol kinetics flow tube. The experimental techniques are all well-tested, appropriate and well-described. The group has a lot of expertise in this area. The key, important aspect of this technique is that ultra trace levels of N* can be measured in the aerosol particles, providing additional observational data that most other experiments (for example those that monitor the gas phase alone) do not.

The main result from this paper is that the observed uptake coefficient for labelled N2O5 onto nitrate particles is much larger than 0.1, which the authors indicate is a measure of the bulk mass accommodation coefficient into aqueous particles being this large in general.

I have a few questions:

1. I am a bit confused by the nomenclature. In Figure 2 (and Equation 3) there is a [N2O5(p)] term. What is meant by that? It is clearly not the dissolved concentration of N2O5 in the particle. Is it instead the concentration of N* in the particle (as stated in line 37)? But this would be orders of magnitude lower than the level of dissolved unlabeled N. So, how do the authors scale N* concentrations to indicate the amount of N2O5 that has been taken up to the particle, which is what I believe this term represents? Also please use either C or [ ] nomenclature for concentration but not both.

2. I must be missing a very important point in the paper, and would like the authors to explain this to make the paper clearer. In particular, I understand that the kinetics experiments with nitrate particles indicate a large uptake coefficient that can be interpreted as a large mass accommodation coefficient. But, I am at a loss to understand why the results with sulfate particles, that show roughly an order of magnitude smaller uptake, cannot also be interpreted as a mass accommodation coefficient measurement. If the reaction proceeds half through the left channel in Figure 3, then the labelled nitrate so formed should stay in solution. If the mass accommodation coefficient is really larger than 0.1 on aqueous droplets then the uptake coefficient in these experiments should have been at least this large. But the observations show much

smaller uptake kinetics. Does this not imply that the mass accommodation coefficient on sulfate must not be as large as on nitrate?

3. The results are interpreted in terms of bulk processes. With very high concentrations of nitrate, could the chemistry for those particles be happening on the particle surface? In this case the kinetics would be surface-uptake controlled and not bulk-controlled, and then this would not be a measurement of the bulk mass accommodation coefficient. Overall, is it possible that the observations are consistent with (fast) surface uptake for the nitrate experiments and with (slower) bulk uptake for the sulfate experiments?

4. It is stated a few times that nitric acid does not volatilize under the conditions of the experiments (and this was the reason to use neutral particles and not acidic ones). Can the authors confirm that this is the case for their conditions?

5. Table 1: Please list how many experiments were conducted per experimental condition.

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