Replies to comments for article "Efficient bulk mass accommodation of N2O5 into neutral aqueous aerosol" by G. Gržinić et al.

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Anonymous Referee #4

1. I'd like the authors to include an estimate of the mixing ratio of N2O5 in the flow tube, as well as HNO3, as well as Va and Sa values (needed in the diffusion correction and to establish the role of possible aerosol size effects in the uptake process).

Response: The aerosol surface area density has already been given in the original table, and the average diameter is mentioned in the text along with the discussion of the diffusion correction.

As mentioned in the reply to question 3 from Referee #2, and question 2 from Referee #3, the design of the experimental setup does not allow to directly assess the levels of HNO_3. As discussed there, for the configuration used in the present setup, we would have lost labelled HNO_3 to the walls efficiently, and based on the measured wall loss rates, the non-labelled HNO_3 levels building up as product would remain low enough to not significantly affect nitrate levels or acidity in the aerosol phase. The gas-phase non-labeled NO_2 and N_2O_5 concentrations were around 10^{11} molecule cm^{-3}. Both aspects will be explicitly mentioned in the revised version.

2. The results hang together well but the low uptake onto sodium sulfate aerosol needs to be discussed in more detail. In particular, the question needs to be addressed as to why is it so low - lower than the reported value by Wahner et al. of 0.037 (+ 0.008 - 0.019)?

Response: As mentioned in the response to the question below, crystallization of the Na_2SO_4 aerosol at 50% is the most likely explanation for the low uptake value measured. The value reported in Mentel et al (1999), which was measured at 71% RH, is consistent with our measurement at 70%. However the measurement done at 50% RH is much closer to the values reported in Hallquist et al. (2003) for dry $(NH_4)_2SO_4$ aerosol. This will be added in the revised version.

3. The aerosol is dried to 40% RH ('to prevent efflorescence' line 39, page 2). This is quite far below the reported efflorescence RH of Na2SO4 of 58%, (e.g. quoted in J. Phys. Chem. A, 2007, 111 (42), pp 10660–10666), so partial or complete efflorescence may explain the low values of uptake observed for the 50% RH case of Na2SO4, as uptake onto solid sulfate is known to be slower. Could this be a possible reason?

Response: Indeed, partial or complete crystallization seems like the most likely explanation, as mentioned by the reviewer, and in light of what is reported by Gao et al. 2007 with regards to the efflorescence relative humidity for Na2SO4. We will update the paper to reflect this.

4. I think the discussion of salting effects requires more detail (do they have any evidence for the salting they propose?) and to be better linked to variation in RH.

Response: within the limited data set of this study, it is difficult to become more conclusive about the salting effects. As mentioned in the discussion, on page 6, from line 7, the fact that the water activity had a stronger effect on the uptake coefficient than different (though still high) nitrate to sulfate molar ratios. As indicated above in response to referee #3, we could only add that Bertram and Thornton (2009) have seen independent evidence for a water activity dependent equilibrium. Also the application of a more detailed kinetic scheme in box model calculations shown in the revised version does not allow constraining the solubility further.

Gao Y., Yu L.E., and Chen S.B., Efflorescence Relative Humidity of Mixed Sodium Chloride and Sodium Sulfate Particles, J. Phys. Chem. A, 111, 10660–10666, 2007.

Mentel, T. F., Sohn, M., and Wahner, A.: Nitrate effect in the heterogeneous hydrolysis of dinitrogen pentoxide on aqueous aerosols, Phys. Chem. Chem. Phys., 1, 5451-5457, 1999.

Hallquist, M., Stewart, D. J., Stephenson, S. K., and Cox, R. A.: Hydrolysis of N2O5 on sub-micron sulfate aerosols, Phys. Chem. Chem. Phys., 5, 3453-3463, 2003.