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

Anonymous Referee #4

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General comments

The authors present results describing an important atmospheric chemical process, the removal of the NOx reservoir by aerosol, and the article contains new data which contribute to the development of physico-chemical models of the uptake process. The experiments are novel in their use of an isotopically labelled N2O5.

Uptake of N2O5 is quantified by the appearance of 13N in the aerosol phase, as determined by measurements of aerosol deposited onto a filter, and this quantity is used to infer the rate of uptake via fits of the ratio of N2O5 in the particle phase to the initial gas phase N2O5 concentration (measured as 13N deposited onto a citric acid denuder) as a function of gas-aerosol contact time. The authors derive gamma(eff) from these fits, which are then corrected for gas-phase diffusion effects, to give true uptake coeffi-

cients, gamma.

The systems studied are sodium nitrate and sodium sulfate aerosol, and aerosols of mixed composition, at 50% and 70% RH and room temperature.

The most surprising result is in the case of the nitrate aerosol. The uptake coefficient for the nitrate aerosol in the range 50%-70% RH has been previously determined to be small, indicating a slow net rate of loss of N2O5 from the gas phase into the aerosol. The authors find the uptake coefficient as determined by the appearance of 13N in the aerosol phase indicates efficient accommodation and reaction with bulk aerosol nitrate. More surprising still, the rate of uptake is larger than previously reported accommodation coefficients, which was derived from the observation that uptake coefficients tend to a maximum value of around 0.04 under all conditions. Thus, the accommodation of N2O5 would appear to be much more efficient than previously considered. This is an interesting result.

The experimental method is well-established and the group have considerable expertise in this area. The paper is generally well-written, figures are clear, and appropriately referenced. While the number of systems studied is rather small, and the paper is consequently rather brief, the difficulty of the measurements and their interest merits publication at this stage. I do feel the paper might be improved with some more quantitative treatment of the kinetics of the uptake process, but would be happy to see the work published at this stage following minor revisions.

Specific Comments

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).

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)?

The aerosol is dried to 40% RH ('to prevent effloresence' 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?

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.

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