

Interactive comment on “Influence of aerosol chemical composition on N₂O₅ uptake: airborne regional measurements in North-Western Europe” by W. T. Morgan et al.

Anonymous Referee #2

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Overview:

Morgan et al describe a new set of N₂O₅ uptake determinations calculated using direct measurements of N₂O₅, NO₂, O₃, NO₃, and particle surface area by employing the steady-state expressions developed by Brown et al. There are few measurements of this kind in the literature and none outside of the continental United States that I am aware of. Previous work by Brown et al. and Bertram et al. have shown that determinations N₂O₅ reactive uptake on ambient aerosol may serve as a unique probe for particle composition and how the organic component of ambient aerosol impacts heterogeneous and multi-phase processes. Morgan et al. find that N₂O₅ uptake was

C6876

efficient (0.01–0.03), perhaps a consequence of high RH conditions sampled here. The authors demonstrate the well-known nitrate effect impacts N₂O₅ uptake, however find little evidence for a suppression from particulate organics as has been observed in previous studies.

In general, I find the paper to be within the scope of ACPD and should be published following the authors attention to the following comments:

Specific Comments:

- 1) It is my understanding that when determining if the steady-state calculation is correct, a model is often used to assess the time required to achieve SS. This model should include the loss rate of nitrate radicals to various VOCs. Was this done? And if so, what measurements were used to assess this and more generally, what is the relative strength of NO₃ reactivity compared with N₂O₅ reactivity in these airmasses? It is important to highlight this comparison given that the data (as shown in Fig. 4) appears to hold some of these answers.
- 2) In equation E3, what surface area was used? An average surface area for the entire flight? An average over the flight leg used to generate the lines in Figure 2? Given that NO₂ (and thus $K_{eq}[\text{NO}_2]$) often co-vary with surface area, it would be important to note how the variability in the surface area impacts this calculation.
- 3) In section 2.4 the authors describe how the ambient surface area was determined using a calculation of the hygroscopicity. It would be helpful to include the values that were used. It appears that the authors used a very small growth factor (similar to fulvic acid). What is the sensitivity of the conclusions to this decision? Given that the retrieved uptake coefficients are already close to the upper limit observed in the laboratory for tropospheric mimics (0.03) a more hygroscopic organic fraction would act to increase the particle surface area and thus decrease the retrieved gamma value.
- 4) Again, with respect to hygroscopicity. How were the organics treated in calculating

C6877

particulate water content for the Bertram and Thornton parameterization? Were they also treated as fulvic acid? How might this decision impact the resulting conclusions?

5) The correlation of gamma N₂O₅ with NO₃ as shown in Fig. 5 is not very strong and appears to be guided by two points. What does it look like as a function of [NO₃-]/[H₂O]I?

6) One of the more interesting aspects of this work is that there does not appear to be a strong correlation between gamma N₂O₅ and organics as has been shown previously. Given that the data set appears to be quite robust and a high resolution AMS was on the airplane, it would be of interest to push this question a bit further. In figure 5, a correlation against the organic mass fraction is shown. Is it possible to expand upon this and look at a correlation with O:C for particles that have similar [NO₃-]/[H₂O]I? This would help shed more light on the role of the organic fraction in suppressing N₂O₅ uptake.

7) What diffusion constant for N₂O₅ in the organic coating was applied for E6?

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