

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 #1

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This manuscript presents an analysis of aircraft observations obtained during the RONOCO campaign in July 2010 over North-Western Europe. The aim was to determine how bulk aerosol chemical composition influences the heterogeneous uptake of N₂O₅. The authors derived the N₂O₅ uptake coefficients based on the method by Brown et al. (2006) using observed mixing ratios of trace gases and aerosol surface area concentrations. These observationally-derived uptake coefficients were then compared to values obtained from various parameterizations in the literature. The best agreement was achieved when using the parameterization by Bertram and Thornton (2009), without considering any effects of chloride or organic coatings, even though

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these species were present in the aerosol.

The paper fits well into the scope of ACP and presents original results: It is the first study to show N₂O₅ uptake coefficients that were derived from observations outside the US. The authors show that current parameterizations of N₂O₅ uptake based on laboratory measurements have difficulties reproducing the observationally-derived values. Documenting such discrepancies is important to eventually improve our abilities to represent these processes in chemical transport models. I recommend this paper to be published, but I do have some questions regarding the data analysis, which are detailed below:

Comments:

1. Equilibrium of NO₃ and N₂O₅: The authors use a box model to check if the equilibrium is established. Why do you not use directly the actual measurements of NO₂, NO₃ and N₂O₅ in a similar fashion to Brown et al. (2003) (see their figure 6)?
2. Aerosol surface area calculation: What hygroscopicity was assumed for the organic material (p. 19683, line 20)?
3. Calculating kN₂O₅: It is not clear from the description how the data was chosen that goes into each fit. This seems crucial for the result. On page 19687, line 20 the authors say: "Case studies were selected during portions of the flight when the aircraft was sampling relatively homogeneous pollution conditions at a constant altitude below 1500m. . .". Can you justify the data selection more quantitatively? Following Brown et al. (2006), the data is separated into regions with distinctly different chemical regimes. A figure similar to Figure 3 in Brown et al. (2006) would clarify this question.
4. Calculating gamma values: page 19687, line 6: How was the uncertainty of 36% derived for gamma?
5. Internal mixture assumption: I wonder how appropriate the assumption is that the aerosol is "internally mixed". This assumption plays into the calculation of the aerosol

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surface area, but also into the calculation of gamma for the population. I agree with the authors that aerosol away from near-field sources appears internally mixed with respect to hygroscopic properties. However single-particle measurements also show that there can be considerable variability in terms of composition. For example, what if the organic material is not evenly distributed over all particles in the population? Wouldn't this lead to a larger spread of gamma values from the parameterizations (e.g. Fig. 6)?

6. Figure 5: Given that there is essentially no relationship between gamma and the organic mass fraction, is there any reason to believe that the organics are mostly water-soluble (i.e. they don't form a distinct coating)?

Typographical errors and other minor comments:

p. 19680, line 16: What is SLR?

p. 19682, line 1: should read "system has to be in steady-state"

p. 19686, line 21: "planetary boundary layer". Should read residual layer.

p. 19687, line 14: the term "gradients" is wrong here. Please rephrase.

Figure 3: Explain in the caption what the boxes, error bars etc. are.

Figure 6: Some axes labels are missing.

References:

Brown et al. (2006), Variability in nocturnal nitrogen oxide processing and its role in regional air quality, *Science*, 311, 67-70.

Brown et al. (2003), Nitrogen oxides in the nocturnal boundary layer: Simultaneous in situ measurements of NO₃, N₂O₅, NO₂, NO, and O₃, *J. Geophys. Res.*, 108, 4299.

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