

Review of “An in-situ flow-tube system for direct measurement of N₂O₅ heterogeneous uptake coefficients in polluted environments”

The authors present a flow tube measurement of the N₂O₅ uptake coefficient that is an extension of the work of Bertram, Riedel, and Thornton. The measurement system is described and it is similar to the earlier design. The main innovations presented here are a more detailed measurement of the residence time distribution in the flow tube and the application of an iterative box model to retrieve the uptake coefficient when ambient concentrations of NO, NO₂ and O₃ are high enough to make 2nd order reactions important in the flow tube. The authors also present ambient measurements of the uptake coefficient which are useful because these direct measurements are rare and limited geographically.

This is an important measurement and should be published in AMT with minor changes.

Suggestion: A method, complementary to the iterative box model analysis, would be to reduce the concentrations of the gas-phase interferers (NO, NO₂, O₃, VOCs) before the N₂O₅ addition using an activated-carbon scrubber that transmits aerosols, such as <http://www.sunlab.com/denuders/>.

Minor issues:

- 1) Typically laboratory measurements of the uptake coefficient on synthetic aerosol are less than 0.04. Although some ambient analyses (Wagner et al. 2013, McDuffie et al. 2018) report uptake coefficients above 0.04 (upto 0.1) for a small subset of the data. It is not clear if these are artifacts of the analysis or real measurements of the uptake coefficient. Here the authors also report a direct measurements of uptake coefficients between 0.04 and 0.1. I would encourage the authors to address the discrepancy between laboratory measurement and their ambient measurements.

If they are real what is aerosol composition? Can the measured uptake be reproduced in the lab with synthetic aerosol?

- 2) It is unclear what parameters were used in the uncertainty analysis. I suspect uncertainty due to the aerosol surface area measurement would be at least +/-25%. In figure 9, there are not smooth exponential decay transitions between filter ON and OFF periods, so I suspect the uncertainty in the N₂O₅ measurement is significant.

On page 8 line 18, please list the key parameters and the uncertainty associated with them.

- 3) Measurements of NO and VOCs are not described. Uncertainty due to reactions of NO₃ with unmeasured VOCs should be bounded.

- 4) The authors show that the residence time in the flow tube is a distribution (ranging over a factor of 2 in residence times), however in the iterative box model only the mean residence time is used. As the iterative box model likely depends in the residence time in a nonlinear way, the author should use a range of residence times in the iterative box model.
- 5) Have the authors measured particle losses in the flow tube? Diffusional and gravitational losses could be important. Could aerosol losses also be RH dependent? If so, please add a few sentences describing the results.
- 6) In figure 9, the periods chosen for analysis seems to be handpicked for stability. If different periods were chosen how would the results change?

Technical issues:

Pg 3, line 26: How does the flow tube pressure relate to ambient pressure?

Pg 4, line 14: how much NO₂ is added with the N₂O₅ addition?

Pg 7, line 4: This sentence is missing a subject

Pg 7, line 20: Please give some more explanation about when non-physical results occur. When the uptake coefficient is small. When aerosol number is low? I expect that in a flow tube with high initial N₂O₅ the box model would work well in most cases.

Pg 8, line 5: please add 'respectively'

Pg 9 line 9: typo 'ere'

Pg 9, line 16: Could you summarize the potential artifacts.

Pg. 10 line 27: missing 'the', 'in aerosol mode'