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Atmospheric Chemistry and Physics Discussions

Interactive comment on “The interaction of N_2O_5 with mineral dust: aerosol flow tube and Knudsen reactor studies” *by* C. Wagner et al.

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In this laboratory study, the authors investigate the uptake of N2O5 to authentic mineral dust, sand, and calcite as a proxy representing part of mineral dust composition in the atmosphere. Mineral dust is one of the major types of atmospheric aerosol. N2O5 is an important NOy species, that acts as a reservoir for NOx. The heterogeneous loss of N2O5 to aerosol particles is one of the most important heterogeneous reaction globally that affects the budgets of O3 and nitrogen oxides in the troposphere. Kinetic data on this reaction are therefore very relevant. Due to significant discrepancies in previously reported kinetics, this new and thorough kinetic study is very welcome and relevant.

While part of the existing discrepancies in kinetic data is due to the way data obtained in Knudsen reactors are being evaluated to obtain uptake coefficients, this study uses

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both an aerosol flow tube and a Knudsen reactor to compare the results for a Saharan dust sample, for which reasonable agreement is reported. The results show that the uptake coefficient is in the lower 0.01 range on dry Saharan dust, which confirms the significant role of this reaction in the troposphere. The sample history indicates a strong role of water on the kinetics, which is not investigated in more detail.

I appreciate the detailed and well thought description of the experiments and their analysis. Overall, the paper is well written and well structured. My only significant concern relates to the way particles are exposed to N2O5 (mode of flow tube operation, see specific comments below). I recommend the paper for publication, if the points below can be adequately addressed:

Specific comments

1) I refer to Fig. 3 showing the response of the N2O5 to pulses of dust particles admitted to the flow tube and the way k_d is derived based on these signals as described on p 13305. The direct comment I have on this mode of operation is that when looking at the transient signals as shown in Fig. 3, I wonder whether the time resolution was sufficient (for detection of particles and N2O5) to fully resolve a quasi-steady state signal for the state, when aerosol is present. The second point relates to a more fundamental issue: Given laminar flow conditions in the reactor as suggested by the authors, I would like to raise the point that the residence time distribution of particles (following a 1/tˆ3 fall off behaviour after half of the average residence time) is not the same as for N2O5, which is essentially 'plug flow' due to radial diffusion. I wonder how long the pulses must be such that the measured drops in N2O5 must not be affected by the inhomogeneity of the N2O5 concentration field in the regions, where the particles travel down along the flow tube, with those in the center travelling quite a bit faster than those closer to the flow tube wall. While the later analysis based on the k_d retrieved as reported do not indicate a problem directly, this might lead to a constant correction, independent of particle number density or injector position.

2) magnitude of correction for diffusion, P13306, L17: because of the non-linearity of the correction I wonder, whether the number would not be different if calculated as weighted average of the correction for each size bin of the distribution.

3) discussion of pore diffusion correction related to Saharan and Arizona Test dusts, P13314 and 13315: I appreciate the thoughtful use of the pore diffusion model. One point to think about might be the following: It seems that for both samples the particle size used to parameterize the porous powders was relatively large. For the Saharan samples it seems to be consistent (on purpose?) with the APS measurements from the AFT. However, this size must not be a primary particle size but rather more a size related to the disintegration efficiency of the brush generator used to suspend the particles. We have shown for ATD (Vlasenko et al., Aerosol Sci. and Technol., 2005) that the powder can disintegrate into submicron particles. Even there, microscopic inspection showed that some of them are agglomerates. Therefore, would a smaller primary particle size lead to different results with the pore diffusion model? This would also indicate that a correction for internal surface would have to be made to the aerosol flow tube results.

Technical comments

P13298, L21: 'which was reported' P13299, L16: missing comma after 'APS (TSI3321)'; and after 'flow tube' on the next line. P13304, L11: differential term should read dc/dt, either brackets should be used to enclose a species name in caps, or just c or other lower cap character to denote concentration. Next line: missing space following the symbol for molecular velocity. P13304, L19: explanation of k_d: I suggest using the term 'gas kinetic collision rate with the dust multiplied by'; rather than 'dust collision rate' P13313, L23: correct to 'considerably larger';

Caption to figure 1: 'RBG' for rotating brush generator Caption to figure 3: 'acquisition'

References

Vlasenko, A., Sjogren, S., Weingartner, E., Gaggeler, H. W., and Ammann, A.: Generation of submicron arizona test dust aerosol: Chemical and hygroscopic properties, Aerosol Science and Technology, 39, 452-460, 2005.

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