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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.

C. Wagner et al.

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We thank Markus Ammann for his comments on our manuscript. His comments and our replies are listed below:

Comment: I refer to Fig. 3 showing the response of the N2O5 to pulses of dust particles admitted to the flow tube and the way kd 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 $1/t^3$ 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 kd retrieved as reported do not indicate a problem directly, this might lead to a constant correction, independent of particle number density or injector position.

Reply: This is an interesting and important point related to axial gradients in the dust number density. The experimental time resolution (i.e. the integration time for N2O5 signal or dust number density) was about 1 s. This is short compared to the duration of the dust pulses, which sometimes lasted several tens of seconds (perhaps pulse is a misnomer in this regard) and was similar to the N2O5 residence time in the reactor. As the reviewer has noted, the width (in time) of the dust pulses varies slightly from the width of the response of the N2O5 signal, the latter being thinner. However, the fact that single data sets with a mixture of sharp dust pulses and broad dust pulses yield (within experimental scatter) the same uptake coefficient confirms that we are not far from steady state. We concede that some of the scatter in e.g. Figure 6 (containing both sharp and broad dust pulses) may indeed be due to such effects. This is however not the main source of error in these experiments. It is true that the residence time of the particles will have a strong radial dependence. Their concentration should however be radially uniform as they are added at large flow rates and velocities at right angles to the main axis of the flow tube, resulting in rapid turbulent mixing. We shall modify a revised version of this manuscript to incorporate elements of this discussion.

Comment: 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.

Reply: In response to this comment, the diffusion correction was carried out using the full distribution and a separate Knudsen number for each bin. The correction factor was

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11.3 % instead of the correction factor of 10.9 % obtained using the method outlined in the text. We mention this in the revised manuscript.

Comment: 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.

Reply: Correct: The SDCV particle size was taken to be the same as the particles observed in the APS, even though they had NOT been processed by the RBG. Scanning electron microscope pictures of the SDCV samples indicate the presence of both large rocks (> 5 microns) and sub micron particles, making the choice of average particle diameter rather difficult. In light of this a detailed re-analysis of the Knudsen data using e.g. different particles sizes in the PD correction is probably not warranted. Instead, we shall modify the text related to pore diffusion corrections and SDCV to emphasise the limitations of using porous, bulk samples of poorly defined grain size.

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. P13304, L11: missing space following the symbol for molecular velocity. P13304, L19: explana-

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tion of kd: 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'

Reply: These corrections will be made as suggested.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 13291, 2007.