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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Enhanced growth rate of atmospheric particles from sulfuric acid" *by* Dominik Stolzenburg et al.

Anonymous Referee #2

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This is important work and therefore needs the best, possible analysis and also a clear presentation of uncertainties. I plan to submit comments on the presentation style and the context of the work at a later date.

(0) Are the particle sizes listed as mobility diameters (see Larriba et al. 2011)? Assumed to be the case for the rest of these comments. . .

0.a) Assuming ammonia is not affecting composition significantly, relative humidities of 38 and 60 % gives bulk densities times wt. fraction of 0.67 and 0.49, respectively. This indicates an expected 30 % difference in growth rates for these two conditions. Is there a humidity dependence in the GRs?

(1) Potential bias in the measurements at the smallest sizes. As an example, the 1.8

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Discussion paper

nm mobility diameter channel corresponds to a 1.5 nm mass diameter.

1.a) If we assume they are large enough to attain bulk properties (and ammonia has no effect) there are about 6 sulfuric acid molecules in this DMA-train channel. Early literature results (Froyd and Lovejoy, Hogan lab) indicate that negatively charged 4 acid clusters can hold very little water thus these small particles will shrink upon charging (since it is likely that 6 acid clusters will also lose much of their water content upon charging.) There is a potential bias in what size to assign to the detection at these small channels: they may actually be a little larger in size than 1.8 mobility diameter before charging. Is it known at what size shrinkage becomes negligible?

1.b) On the other hand, the 1.8-2.2 nm DMA-train signals may come from neutral particles that have not attained enough water to reach bulk values. Yet there will be a size at which bulk properties are better achieved, say 3.2 nm for argument. Then between the small-diameter channels and the 3.2 nm diameter (or larger) there will be swelling due to water uptake. This is an effect that is not directly related to collisional rate coefficients.

These two effects would affect the measured GRs in the same direction it seems.

(2) Confusion about Figure 2. It seems that the monomer is plotted with mass diameter (assuming bulk hydration and density) while the DMA-train results are plotted as mobility diameters? Secondly, where is the 1.8 nm data? This size might be especially sensitive to evaporation. Thirdly: it is not clear whether an additional first-order loss term due to evaporation would appear as a negative or positive deviation from the sum of the calculated/estimated first-order loss rates (i.e. wall loss, coagulation, and dilution): for example, a 2.55 nm particle evaporating might cause a bump up in the 2.2 nm channel's signal (depending on relative abundances etc.)

(3) The smallest channels' appearance times may be most affected by the time it takes H2SO4 to reach steady state. How has this been addressed?

(4) Are not the GRs subject to a -50/+33 % systematic uncertainty due to the +50/-33% uncertainty in measured H2SO4? This should be reflected in the figures and particularly in Fig. 4b where it is stated that systematic uncertainties are represented by the pink band.

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