Atmos. Chem. Phys. Discuss., 8, S4921–S4923, 2008
www.atmos-chem-phys-discuss.net/8/S4921/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol dynamics simulations on the connection of sulphuric acid and new particle formation" *by* S.-L. Sihto et al.

Anonymous Referee #2

Received and published: 16 July 2008

Sihto et al report on aerosol dynamics simulations in order to investigate how the relation between H2SO4 and particle formation rate and concentration changes as they grow from nucleated size to 3 nm. They used the activation mechanism and ternary nucleation as the two candidates for nucleation, and then compared the simulation exponents (J ~ [H2SO4]exp) to those found in selected atmospheric measurements.

Not surprisingly, the activation mechanism produces smaller exponents for J3 and N3 than ternary nucleation. Therefore, and in light of the known exponents (for J1 at least) for the different nucleation mechanisms (1 for activation, 2 for kinetic, and substantially larger for ternary nucleation), I don't understand why the authors didn't include kinetic nucleation in their simulations, perhaps even instead of ternary nucleation. The conclusion that the ternary nucleation mechanism doesn't provide a similar exponent as the

measurements is hardly surprising. Generalizing that conclusion, while other studies found that the ternary nucleation parameterization produces good results in a different location, is not warranted. I suggest running simulations using kinetic nucleation, since that is a candidate with some promise to provide similar exponents as the measurements under consideration, and it would be interesting to compare those results with activation nucleation.

Specific comments:

P 11375, on estimating the time delay: In the text (lines 7-10) it is mentioned that the time delay is estimated visually rather than by investigating what resulted in the maximum correlation, but caption of Fig 3 mentions that the time delay of 1.5 hours corresponds [to] the best correlation. Looking at Fig 3 a and b, I fail to see that the correlation would be independent of the time delay, as is implied by the text. If I misunderstand something here, then please clarify. The labels seem to be mixed up for N3-6 and H2SO4 in either fig 3a or 3b.

P 11376. Size dependence of the growth rate: Based on the text, I assume that fig 5 is for csat = 10e6 (please mention this clearly). Fig 6 however makes me think differently, or are the labels mixed up? (The text mentions that the minimum in the growth rate disappears when csat goes to zero.) All in all, this is very confusing. The size dependence of the growth rate seems to have a significant effect on the resulting exponents for N3 and J3. It is therefore important to provide the reader with some sense of the uncertainties involved: How dependent is the size dependence on other factors besides csat? How dependent are the exponents on different factors than csat? The authors mention oligomer[iz]ation as a candidate to explain the low apparent value of csat. Are there other possibilities that could also explain the results? Different concentrations of H2SO4 and/or organics for example? Or different values describing the molecular behavior in equations 1 and 2? I would like to see the discussion expanded here.

P 11379 regarding size and composition of the critical cluster: The critical cluster in

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ternary nucleation is expected to consist of 5-6 H2SO4 molecules (corresponding to the exponent value) and to be around 1 nm diameter. This raises the question what the critical cluster exists of in activation nucleation, when according to the exponent it would only consist of 1 H2SO4 molecules, but is likely 1.5 to 2 nm in diameter?

P 11380, line 13-15: This conclusion is too strong and generalized, see also the comment of referee 1.

Abstract line 13-16: mention to which nucleation mechanism these results apply.

Please reword P 11367 Lines 22-25

Define peq, org (eqs 1 and 2, p 11368)

P 11370 line 21: "These studies have considered" to make it clear that you refer to the just referenced studies.

P 11371 line 1-2. I appreciate the attempt to put these equations in words, but it would be more helpful if the symbols are then described in words as well, otherwise it fails the purpose.

P 11372 line 7. Using equation 7 (instead of 6). One equation (before eq 7) is unnumbered.

Figures 2-4 and 7 are too small, especially the axes labels and legends.

P 11376, line 3: In field measurement data [from the Boreal forest], etc

Here and there the language needs to be corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 11363, 2008.

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