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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric homogeneous nucleation of H_2SO_4 and H_2O" *by* D. R. Benson et al.

Anonymous Referee #1

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The authors replies to my comments are unsatisfactory on a couple of crucial points:

1. The authors 'justify' having un-quantified inpurities in their system by the fact that other previous research have had the same 'problem' also. I feel that every scientific paper should have some important new information that adds to the existing knowledge. When looking into new things (as the first one, or among the first) it is ok/natural to be 'not perfect' (have uncertainties, impurities etc.). However, as progress is made, these uncertainties are typically decrasing (or hopefully removed altogether) - and progress is made. This is not the case here.

2. Still, the authors present a set of (possibly) valuable experimental data. It should be, however, much more thoroughly quantified/analyzed. My suggestion is to spend much more time on the growth rate. Now, the authors' response to my criticism of having a

factor of 28 unknown cause in the growth rate is, again, 'other researchers have had uncertainties also' and 'only a limited number of growth theories exist' (which is an odd statement). Condensation-theory is much more 'well established' than throries on nucleation - of course there are uncertainties related to thermodynamical quantities, however. In any case, for the ternary system of water-sulfuric acid-ammonia, it should be possible to estimate growth rates for specified conditions and concentrations. The same, apparently (by the references mentioned in the nice comment by Jianxin Ma), is true also for amines. So why not play with condensational growth models and see what kind of ammonia/amines concentrations (or are other significant concentrations of 'impurities' needed?) are needed to explain the observed growth rates - and compare them with e.g. the detection limits of the instruments.

3. The main value of the Berndt et al. and Sipila et al. papers is the fact that both works carefully try to find out possible causes for the different slopes in the logJ-logH2SO4-plots. The explanations on effects of detection limit and residence time are easy to grasp and clear now. One other thing (removing much of the effect of growth) is using new devices with low detection limits, so that measurements gradually approach 'nucleation size'. Since here a 3nm-cutoff CPC is used, any conclusions on the slopes are affected both by a) a longer range to cover by growth and b) other vapors participating on both nucleation and growth.

4. I still also protest the fact that the authors refer to binary nucleation at all, when writing about these experiments. For many previous works (in which there has been other participating vapors also) this is ok because at the time the researchers did not know any better. Now the situation is different.

5. (minor point) When I asked for more reasoning about the 'threshold' of 1.e+6 for particle formation, I meant how (in detail) it is seen from the results presented in this manuscript - so please write a coule of lines of advice to the readers what to look for in the figures...

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 29051, 2010.

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