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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.

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I am a chinese phd student, currently i am working on a project regarding atmospheric nucleation (focus on the effects of ammonia and amines). Not sure whether my comments are useful, but i would like to point out that:

RE: Dr. Dave Benson is not immediately available because of personal circumstances and as co-corresponding author, I am responding to you on his behalf. Your comments on ammonia and amines are important. We would also like to draw your attention to our two ACPD papers [Benson et al., 2010b; Erupe et al., 2010], in which we have investigated how ammonia and trimethylamine can affect sulfuric acid nucleation. Your suggested three papers were cited in [Erupe et al., 2010]. With regard to your first two comments below, briefly:

a. We have used certain Teflon material in our system, instead of stainless steel which is most susceptible for ammonia contamination [Benson et al., 2010a; Nowak et al., 2002; Nowak et al., 2006; Nowak et al., 2007; Yokelson et al., 2003] to reduce contamination of NH3. There are also other technical efforts in this regard, as detailed in Experiments section.

b. We have also used CIMS to measure directly how much ammonia and trimethylamine existed in the system as impurities (Experiments).

c. Whether these impurities can affect BHN or not is unclear. But from our THN studies with ammonia and trimethylamine, it seems that sub-ppbv level of base species have very moderate effects on nucleation [Benson et al., 2010b; Erupe et al., 2010]. We only see significant effects with high NH3 (tens of ppbvs) [Benson et al., 2009]. Additionally, even if the system has multiple chemical species, we cannot simply assume that nucleation would take place through multicomponent processes; rather this depends on several conditions including how much we have these ternary species in the system, for example, and sometimes, nucleation can take place solely via BHN even in the presence of ternary species [McGraw and Zhang, 2008], which certainly does not exclude the case of these ammonia or amine impurities.

(1)The role of ammonia in the nucleation should always be paid careful attention. Indeed, impurities of NH3 are hardly to remove. So a clear explaination and treatment to ensure a contition for binary nucleation is necessary.

(2)on the other hand, the effects of amines might be much more important.

Recently, I found a couple of important papers, probably worthy to be mentioned.

Lin Wang, Alexei F. Khalizov, Jun Zheng et al. Nature Geoscience 3, 238 - 242 (2010) (Nanoparticle growth increased with relative humidity in the presence of glyoxal and trimethylamine)

X.Ge, A.S.Wexler, S.L.Clegg. Atmospheric amines – Part I. A review Atmospheric En-

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vironment. doi:10.1016/j.atmosenv.2010.10.012 (Sources of a lot of amines are summarized, clearly shows that amines have similar sources as NH3, probably can be referred as NHx to indicate NH3 and amines)

X.Ge, A.S.Wexler, S.L.Clegg. Atmospheric amines – Part II. Thermodynamic properties and gas/particle partitioning. (I am communicating with the authors why they didn't present the results regarding aminium sulfate/bisulfate while there are results or chlorides and nitrates. At least, they showed that amines typically have a stronger partitioning ability into the particle phase than ammonia based on equilibrium thermodynamics.)

(3)I am wondering Why 287K is chosen to be "atmospherically relevant conditions"?

RE: This is a temperature that falls into the typical lower tropospheric condition.

4) D. Brus, et al. Homogenous nucleation of sulfuric acid and water at atmospherically relevant conditions. Atmos. Chem. Phys. Discuss., 10, 25959-25989, 2010.

RE: We do plan to discuss this paper, which came out within the same week as our current manuscript.

This MS almost talked about the same question as this paper. The authors should compare with each other.

Thanks a lot Jianxin

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Benson, D. R., et al. (2010b), Laboratory Observations of Ternary Homogeneous Nu-

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Erupe, M. E., et al. (2010), The effect of trimethylamine on atmospheric nucleation involving H2SO4, Atmospheric Chemistry and Physics Discussions, 10, 27673-27693.

McGraw, R., and R. Zhang (2008), Multivariate analysis of homogeneous nucleation rate measurements. Nucleation in the p-toluic acid/sulfuric acid/water system, J. Chem. Phys., 128.

Nowak, J. B., et al. (2002), Chemical ionization mass spectrometry technique for the detection of dimethylsulfoxide and ammonia, J. Geophys. Res., 107(D18), 4363, doi:4310.1029/2001JD001058.

Nowak, J. B., et al. (2006), Analysis of urban gas phase ammonia measurements from the 2002 Atlanta Aerosol Nucleation and Real-Time Characterization Experiment (ANARChE), J. Geophys. Res., 111, D17308, doi:17310.11029/12006JD007113.

Nowak, J. B., et al. (2007), A chemical ionization mass spectrometry technique for airborne measurements of ammonia J. Geophys. Res., 112, D10S02, doi:10.1029/2006JD007589.

Yokelson, R. J., et al. (2003), Evaluation of adsorption effects on measurements of ammonia, acetic acid, and methanol, J. Geophys. Res., 108(D20), 4649.

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