Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-800-SC1, 2020
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurement Report: Sulfuric Acid Nucleation and Experimental Conditions in a Photolytic Flow Reactor" *by* David Roy Hanson et al.

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Received and published: 7 October 2020

In this short comment we provide evidence that the referee has reasonably requested. In one of their points they asked for a comparison of our model to the nucleation rates from the ACDC model published in Kuerten et al. [2016].

With the thermodynamics set to those of Ortega et al [2012] (with corrigendum for 4a, 3b cluster) and letting clusters containing 5 sulfuric acid molecules accumulate, we calculated the nucleation rate of 5 acid clusters. This appears to be the nucleated-particles / clusters used for the ACDC model as well (might be 5a and 4b and larger). Please see Hanson et al. [2017] (DOI: 10.1021/acs.jpca.7b00252) section 2.4 and in

its supplement S2.1 and S2.4 for additional information on the 0-D (box) model.

Using the 278 K data presented in Kuerten et al. [2016] for 100 pptv NH3, we simulated three points along the neutral ACDC line (green solid line): SA/cm-3 and J/(cm-3 s-1) of (i) 9e6 and 1, (ii) 1.8e7 and 10, and (iii) 3.8e7 and 100. The results are Jbox/(cm-3 s-1) = 1.2, 12 and 112, respectively. The agreement is good. (*)

We must point out though that using Ortega NH3-H2SO4 thermodynamics limits the simulations to a maximum of 5 acid and 5 base molecules which is problematic for many experimental conditions, particularly for simulations warmer than 278 K. For simulations at 292 K and 100 ppt NH3 and SA at 1e8 cm-3, 5a 5b cluster nucleation is 10.1 cm-3 s-1 which agrees with ACDC as presented in Kuerten et al. If a reasonable extrapolation of the Ortega thermodynamics is applied to the 5a 1-5b clusters such that 6a 1-5b clusters are the accumulation clusters (i.e. the 5a clusters are allowed to evaporate a and b molecules: see plot below) the nucleation rate falls to 1.6 cm-3 s-1. In fact, that is one of the main points of our Measurement Report: that the thermodynamics of the clusters up to ten acid molecules are needed in the NH3-H2SO4-H2O system, even at 278 K - see (*) below.

(*) At 278 K, evaporation rates of the 5a clusters are important. If a reasonable set of free energies are used for the 5a 1-5b clusters, they can evaporate in the model with the accumulation clusters set to the 6a 1-5b clusters. The box model nucleation rates decrease by 32, 64 and 86 % for the (i) through (iii) scenarios, respectively. The trend continues as the thermodynamics are further extrapolated and the accumulation cluster size increases.

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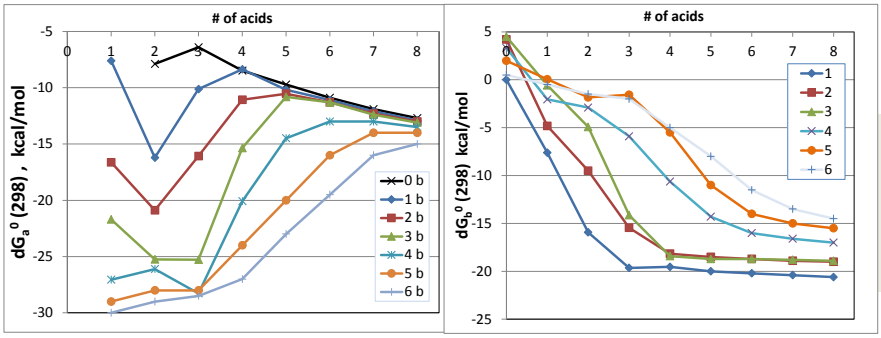

These are step-wise standard Gibbs energy changes for acid (left) and base (right). Number of base molecules indexed by color.
Any values for 5a or larger or 5b or larger are extrapolated from the Ortega et al. [2012] free energies.

Fig. 1. Free energies.