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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.

Anonymous Referee #1

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The measurement report by Hanson et al. (2020) is a follow-up publication on their paper from 2019 (Hanson et al., 2019). In both publications, a photolytic flow reactor (PhoFR) is used to generate H2SO4 from HONO photolysis and subsequent reactions with SO2, O2 and H2O. For concentrations of H2SO4 in the ~1e+09 cm-3 range and a reaction time of ~5 s, new particle formation for the binary H2SO4-H2O system occurs. The H2SO4 concentration is calculated from a chemistry model described by Hanson et al. (2019), with a model update in the current study. The newly formed particles are measured with a SMPS (Scanning Mobility Particle Sizer) using a CPC (Condensation Particle Counter) with DEG (diethylene-glycol) as the condensing liquid. This system can measure the particle size distribution starting at diameters of ~1.5 nm

(Jiang et al., 2011). In the current publication a second TSI ultrafine CPC, modified according to Kuang et al. (2012), measures the total particle concentration. From the particle measurements formation rates are derived as a function of the H2SO4 concentrations. Besides the binary system, further measurements are presented by adding different amounts of NH3. Another set of experiments investigates the dependency of the new particle formation rates on different RH settings. Compared with the previous publication, the current study presents several important new upgrades and results: (1) The cleanliness of the PhoFR has improved. This lowers the baseline particle concentrations for the nominally pure binary system. This is important because contaminants (e.g., NH3 or amines) tend to influence nucleation experiments especially at the warmer (room) temperatures. (2) The effect of NO from the HONO source was included in the chemistry model to calculate H2SO4, which should lead to more accurate sulfuric acid concentrations. (3) An ultrafine condensation particle counter is used to cross-check the numbers from the DEG-SMPS. The new findings yield a revised set of thermodynamic data for the calculation of new particle formation rates (NPF) in the binary and the ternary system. These chemical systems are globally important for NPF. Overall, I recommend publication of the manuscript by Hanson et al. after they have addressed the comments listed below.

Comments

(1) Section 2: Although the chemistry model is described in the earlier publication by Hanson et al. (2019) it would be good to add a paragraph, which summarizes the chemistry treated by the model.

(2) It is mentioned that the binary nucleation experiments yield the lowest values reported so far. The authors should include a figure, where all their measurements (the earlier ones from 2019 and the current ones) are inter-compared with the results from other studies. Currently such a figure is only shown for the experiments with ammonia but not for the nominally binary system.

(3) In Figure 7 results from a nucleation and growth model are shown for different sets of thermodynamic data. This model is probably rather complex and therefore evaluation would be beneficial. Evaluation could be performed by using an identical set of thermodynamic data and compare the model output to another model. This could, e.g., be done for the ACDC (Atmospheric Cluster Dynamics Code) model together with the thermodynamic data for H2SO4-NH3 nucleation from Ortega et al. (2012). Results for these thermodynamic data using ACDC were presented by Kürten et al. (2016).

Further comments

L155 (page 5): Please specify why NO accelerates the H2SO4 production?

L282 (page 9): Why was the CPC inlet exposed to room air?

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