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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Enhanced growth rate of atmospheric particles from sulfuric acid" *by* Dominik Stolzenburg et al.

**Anonymous Referee #3**

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This manuscript (acp-2019-755) describes measurements of <10-nm diameter nanoparticle growth rates from sulfuric acid nucleation under well-controlled, low-ammonia conditions in the CLOUD chamber. Analysis of the growth rates indicates that nanoparticle growth proceeds at a rate faster than the hard-sphere collision rate for sulfuric acid condensation. The explanation for this enhanced growth rate is due to an enhancement in the sulfuric acid collision rate from dipole-induced dipole interactions between the vapor molecules and nanoparticles. Consequently, smaller particles exhibit enhanced growth rates relative to larger particles. Incorporation of these enhanced growth rates for the smallest particles into a global model increases the predicted particle number concentration by 50% in the upper free troposphere, demonstrating a potential global impact.



This is a well-written manuscript describing important results. The manuscript is within the scope of Atmospheric Chemistry and Physics, and will be suitable for publication once the comments below, along with those of Anonymous Referee 2 and Christopher Hogan, are fully addressed.

Comments:

1. Both Anonymous Referee 2 and Christopher Hogan raise significant concerns about the diameter values (mobility vs. mass) used in this study and how those values impact the experimental analysis and conclusions. This reviewer agrees with these comments but will not reproduce them here. The comments of Anonymous Referee 2 and Chris Hogan must both be addressed in revision. Changing the particle diameter used in the analysis may also impact the comparison to Lehtipalo (2016).

2. This manuscript would benefit from providing additional context with respect to reconciling measured nanoparticle growth rates to gas phase sulfuric acid concentrations. Significant effort was devoted to developing and applying models that provide closure for nanoparticulate and gas phase sulfuric acid (see work from McMurry, Smith, and Johnston, e.g. Kuang et al., 2010, 10.5194/acp-10-8469-2010; Smith et al., 2008, 10.1029/2007GL032523; Bzdek et al., 2013, 10.1039/C3FD00039G). While these studies looked at somewhat larger particles (∼5-20 nm diameter) during field studies, the main conclusion was that sulfuric acid addition to ambient nanoparticles proceeds approximately at the collision limited rate.

3. Figure 1b: What is the cause of the deviation from linearity at high [H2SO4]?

4. Figure 4: The explanation of the enhancement factor for the charge-dipole interactions (blue dotted line) should be clarified. The caption states that the blue lines are enhancements relative to the hard-sphere limit (black dotted line). If that is the case, then would not the blue-dotted line be obtained by dividing the red dotted line by the black dotted line in Fig. 4a? Instead, it appears the blue dotted line arises from dividing the red dotted line by the red solid line (dipole-induced dipole interaction).

5. Figure S1c: Is there a factor of several orders of magnitude (e.g. 107) missing from the axis label for sulfuric acid?

6. Figure S3: Please clarify what is meant by “the effect of hidden sulfuric acid”.

7. Figure S4: Are the labels on this figure correct? The figure caption states that the yellow bars indicate water content, but the label in the figure only concerns sulfuric acid.

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