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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Total sulphate vs. sulphuric acid monomer in nucleation studies" *by* K. Neitola et al.

Anonymous Referee #1

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The authors have used a thermally controlled saturator to produce sulphuric acid and measure the sulphuric acid monomer concentration using CIMS and CI-APi-TOF, and the total sulphate concentration using MARGA. The concentration measured by MARGA is observed to be 1-2 orders of magnitude higher than the concentration measured by the CIMS and CI-APi-TOF. Theoretical predictions of the sulphuric acid vapour concentration are found to agree well with the observations by MARGA.

The observed discrepancy in measured concentrations between the instruments could be of importance for the scientific community. However, the way it is communicated in this manuscript I cannot recommend it for publication. There is not any explanation for the observed discrepancy until the conclusions section (which looks more like a discussion section in the current state). In that section, the authors mention the possibility of

sulphuric acid forming clusters with contaminants like e.g. ammonia. That is a possible explanation but to prove that it would have to be measured and quantified. And even then, the authors would have to motivate why they think that would be an important observation (like e.g. a higher fraction of sulphuric acid-base clusters than expected). In the abstract, there is nothing written about possible explanations for the discrepancy.

The authors seem to have done a good job in performing their measurements and in evaluating the importance of potentially important factors like e.g. relative humidity, wall losses, and flow rates on measured concentrations. However, since the reason for the discrepancy in the measured concentrations is not known there is no clear message in this manuscript. The manuscript is generally unfocused, and it is unclear what the actual goal is of the study. When introducing the study in the introduction the authors write: "Here we present a way to produce sulphuric acid vapour from thermally controlled saturator in a wide range of sulphuric acid concentrations". Reviewer 2 from the review of this manuscript in ACPD in 2013 pointed out that the use of an H2SO4 saturator is not new" and the authors seemed to agree on this in their response. Still one gets the feeling when reading both the introduction and conclusions sections that this method of producing H2SO4 is one of the main points of the paper. In addition, the language would have to be checked by a native English speaker.

Other comments:

1. The motivation of sections 3.3-3.4 is vague. The authors should spend more effort in motivating why they compare their formation rates with Brus et al. rather than presenting a lot of figures and describing what they show. There seems to be no important lesson to be learnt here, or at least it is not communicated well enough.

2. Figure 5 shows how the particle number concentration and diameter change as the sulphuric acid monomer concentration increases. The sulphuric acid concentration is within the range of typical atmospheric concentrations, so what is the reason for the rapid growth when the vapour concentration increases? In the atmosphere, growth

rates are normally a few nm per hour and organics do most of the job. Here the residence time is only 30 s.

3. Is there a reason for the total sulphate concentration measured by the MARGA (saturator only) being higher in Fig. 4 (2*109 at 280K) than in Fig. 3 (1*109 at 280K)?

Some of the typing errors:

Page 25788, line 2 of the abstract: “a crucial factor”.

Page 25789, line 26: “the evaporation method”.

Page 25790, line 2: “The SO2 oxidation”.

Page 25790, line 18: “a thermally controlled saturator”.

Page 25790, line 19: “concentrations”.

Page 25798, line 24: “An inlet pipe”.

Page 25797, line 9: “fits”.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 25787, 2014.