

Interactive comment on “Growth of sulphuric acid nanoparticles under wet and dry conditions” by L. Škrabalová et al.

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We thank the reviewer for the helpful comments. Please find our point-by-point responses below.

Referee 1:

1. The main findings listed in the conclusions (as I see them) are: 1. The results are reproducible. 2. High acid concentrations result in higher growth rates than low acid concentrations. 3. Ammonia promotes particle growth. 4. Sulphuric acid alone cannot explain the growth rates. 5. The wall losses cannot be described by the correlation suggested by Hanson and Eisele (2000) and are substantially lower. Equilibrium conditions between the gas and wall are suggested. 6. Wall losses are important in nucleation

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experiments, since the real amount of sulphuric acid is important to know. So what is new information here??? This question is especially important since the 'companion paper' by Neitola et al. seems to describe the same system and also look at the important question in which form the sulphur is in the particles. I have a hard time to find in the paper the scientific novelty, even if the topic itself is hot. Perhaps the key interesting finding of the paper is presented in the abstract, that the effect of RH on growth was found to be inconsistent: at low acid conc. high RH seems to slow growth but at high acid conc. a higher RH increases growth. This observation is, however, not explained.

We agree with general conclusions as seen by referee, however we see the added value in quantification of growth rates of newly formed particles. Also as pointed out by the second referee, very few laboratory studies have been done to investigate the growth of newly formed particles and the role of sulfuric acid in the growth dynamics. Therefore our work provides very relevant findings comparable with observed growth of particles produced by atmospheric new particle formation and in laboratory experiments. We agree to referee's opinion that inconsistent effect of relative humidity on growth rates deserves closer attention and this topic will be discussed in more detail in the revised version of the manuscript.

2. In my view the key problem of the paper lies in the poor quantification of the wall losses, which is partly due to the unknown impurities in the system - and affects all further analysis. The presence of impurities (e.g. ammonia) in the system, even at low concentrations, will probably have significant effects on the aerosol dynamics of the system. As a significant fraction of the sulphuric acid may be in larger clusters (as studied by the same group in Neitola et al.), it can have big effects on both the growth and the deposition behaviour of the aerosol. In addition, the presence of impurities on the wall may also affect deposition rates. Partly because of these reasons it might not make so much sense to analyse whether the deposition expression by Hanson and Eisele, that has been developed for a different system, works or not? In addition, the statement "We estimated the real wall losses of H₂SO₄ by matching the predicted and

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observed growth rates through tuning the initial H₂SO₄ concentration in the model", on page 24103, will probably result in wrong wall losses because of these same reasons? With these uncertainties in wall losses and not knowing the impurity concentrations (or more details of the cluster distributions), much of the growth rate data is unfortunately not very interesting and is probably very system specific. As the group seems to have access to all the newest toys (CIMS, APiTOF, MARGA...), as shown by the much better characterized experiment by Neitola et al., why were they not applied here?

The aim of the current work was to study the growth of newly nucleated particles under different experimental conditions and the role of sulfuric acid played in the growth. The detailed investigation of the presence of impurities and their role in the overall process is beyond the scope of this study. The concentrations of ammonia in the system was estimated based on the data from Neitola et al., (2013) who used the same setup in their experiment. As mentioned in the manuscript (page 7, line 7 and 8), the estimated average NH₃ concentrations were 60 pptv under dry conditions and 126 pptv under wet conditions, those values were measured several times in the same system and are well reproducible. Also the levels of ammonia are very close for dry and wet conditions to that observed by e.g. Benson et al.(2011) below 100 pptv for RH from 6–40 % in completely different experimental setup. Due to presence of ammonia in the system, our model accounts for three different degrees of neutralization of formed particles by ammonia. All three model setups provide similar growth dynamics in terms of magnitude. As stated in the manuscript, we can not rule out the presence of additional chemical compounds in our system. Even though the impurities like amines and other organics were measured by MARGA in the same experimental setup several times (see Neitola et al, 2013), their levels were always below experimentally determined detection limits (for amines ~100 pptv) . The discussion on their possible role in the growth process will be added. To address the estimated wall losses, the referee is right that using the deposition expression by Hanson and Eisele(2000) in our study does not provide correct results due to wall-equilibrium conditions in the flow tube. We did not measure the sulfuric acid concentration directly during experiments. Additional calculation were

therefore performed in order to quantify the wall losses incorporating the wall loss rate coefficients of sulfuric acid determined in previous study (Brus et al., 2010) into the model. The model evaluation and discussion about the wall losses will be added to the revised manuscript.

The authors of this manuscript share only two authors from extensive author list of Neitola et al. (2013), and the group does not possess "all the newest toys": for the authors of the current manuscript, the access to these instruments is very expensive and limited to short campaign studies. However the knowledge from previous studies like Neitola et al., 2013 is applied and throughout discussed in this manuscript.

3. Acceptance of this, in my view, would require better quantified wall loss rates and cluster distributions and some idea of the impurities (and their role in the dynamics). If this is not possible, I encourage the authors to merge the key findings of the paper with Neitola et al, if possible.

As noted in the response to the second comment, additional modelling was performed and discussions regarding the wall losses, impurities and their role in the dynamics will be added to the revised manuscript. Investigation of cluster distributions was beyond the scope of this study, as no measurement of clusters was performed during experiments and such data are thus not available. Therefore we are not able to give any statement regarding the cluster distributions as a result of this study.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 24087, 2013.

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