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Interactive Comment

# *Interactive comment on* "Growth of sulphuric acid nanoparticles under wet and dry conditions" *by* L. Škrabalová et al.

**Anonymous Referee #2**

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This discussion paper describes studies of sulphuric acid particles nucleating and growing in a laminar flow diffusion tube. The experiments are performed using varying residence times in the flow tube and in several sulphuric acid concentrations and relative humidities. The total particle number concentrations and number concentration size-distributions are determined for the steady-state conditions in the flow tube. The experimentally determined particle growth rates are compared to model calculations that take into account hygroscopic growth of the particles as well as their neutralization by ammonia, which is assumed to be present in the system at contaminant levels.

As sulphuric acid is one the key species in the nucleation of atmospheric particles, reports of controlled laboratory studies giving information on its role in the growth process are very relevant. In the current literature there are not many reports of aerosol



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Discussion Paper

growth rates from laboratory studies. Therefore, this discussion paper is suited for final publication in Atmospheric Chemistry and Physics, but I would like the authors to make clarifications on key issues related to both the experiments as well as the subsequent analysis of the data.

General comments:

1) Please describe the experimental setup in more detail, otherwise it is very hard to understand the results of the experiments based on this paper alone.

2) This study is based on sulphuric acid concentrations that are calculated using a formula for the temperature dependent saturation vapor pressure. How accurate are the concentrations calculated using this formula? The authors should discuss about comparison of the calculated concentrations to available measured sulphuric acid concentrations. If the measurement setup is the same as the one used by Neitola et al. (2013), authors could refer to this study for comparisons.

3) Are the particle growth rates calculated assuming that nucleation occurs at the very beginning of the flow tube? If this is the case, has it been studied whether particle nucleation occurs also some distance along the flow tube, and what effect this would have on the calculated particle growth rates? What about the condensational loss of sulphuric acid to the particle phase, can this process be neglected when considering the sulphuric acid concentration inside the flow tube?

4) Are the particles assumed to grow with constant concentrations of sulphuric acid, water and ammonia in the flow tube? Is there any measurements or modelling studies about the vapor concentration profiles along the tube? How would the modelled growth rates change if the possible profiles of the condensing vapors were taken into account? Now the authors explain that the best agreement between the observed and modelled growth rates is achieved by using the calculated sulphuric acid concentration at the beginning of the flow tube. If the sulphuric acid concentration is decreasing along the flow tube, this would make the modelled growth rates an overestimate. Also, the



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authors choose not to take into account loss of sulphuric acid vapor to the tube walls, as this provides best agreement between modelled and observed growth rates. However, there are other possibilities to make the modelled results match the observations, and I would recommend the authors to include discussion on those.

5) Have the authors considered taking into account the modifications proposed by Lehtinen and Kulmala (2003) and Nieminen et al. (2010) to the mass transfer equation (Equation 4 of the current paper)? For particles smaller than about 10 nm it was shown by Nieminen et al. (2010) to enhance the growth rates by upto a factor of two for vapors with molecule masses in the range 100 – 150 amu (for example ammonium sulphate which is considered in this study).

Specific comments:

Chapter 2.1, Equation 3: Please give also the range of the sulphuric acid diffusion coefficient variation between dry and wet conditions.

Chapter 2.2, line 28: Is the initial particle diameter the size of the nucleated critical cluster? How well is it known at which point of the diffusion tube the particles are nucleated? This affects the residence time in the tube and therefore the growth rates calculated by Equation 7.

End of Chapter 2.1: In the last paragraph of Chapter 2.1, Equation 7 is used to calculate the particle growth rate. It should be given more clearly what is the initial particle diameter dinit used in the calculations. The growth time for the particles is taken as the residence time of the sample air travelling through the flow tube. Does this mean that the particle formation is assumed to occur right at the beginning of the flow tube? Is there any evidence that this really is the case, or whether the formation occurs at some length into the tube? This would directly affect the calculated growth rates. Is there any information about the sulpuric acid concentration profile along the flow tube, if it is constant or decreasing along the tube? This would also affect the particle growth as they are moving along the flow tube. Could the uncertainties on the calculated growth



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rates caused by these effects be estimated?

Page 24101, end of Chapter 3.3: Could the changing behaviour of growth factors in higher sulphuric acid concentrations be explained by a larger fraction of acidic (and therefore more hygroscopic) particles at higher H2SO4?

Chapter 3.4: In Neitola et al. (2013) describing experiments using the same setup, wall losses were estimated to be similar as in this study. However, in this study the authors conclude that the wall losses are estimated to be too high, or do not correctly take into account the sulphuric acid concentrations the particles are exposed to during their growth in the flow tube. Therefore, the authors have chosen to neglect the wall losses entirely in this study. Since this is very crucial assumption regarding the conclusions of the paper (namely agreement between the modelled and observed growth rates), the validity of neglecting the wall losses should be discussed in more detail. Also, what would be the implications of neglecting the wall losses for the Neitola et al. (2013) study? Is there any information about the sulphuric acid concentration profile along the flow tube, or could this be estimated? The particles are growing the whole time they are travelling through the tube, and if there are differences in the concentrations of the condensing vapors along the tube, this affects the particle growth rates. Assuming that the particles grow the whole time with the rate determined by the vapor concentration at the start of the tube is probably an overestimation.

Technical comments:

Page 24105, line 3: Consider revising “A trend line which presents the theoretical predictions of the growth of ...” into for example “The theoretical predictions of the growth of ...”.

Page 24105, line 15: In reference to the iodine species, the reference for O’Dowd et al., (2002) listed in the reference list is probably incorrect, should be O’Dowd et al. (2002): Marine aerosol formation from biogenic iodine emissions, Nature, 417, 632–636. Also, the reference to Kulmala et al. (2013) is probably misplaced here, it does not discuss



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condensation of iodine species.

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