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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Total sulphate vs. sulphuric acid monomer in nucleation studies: which represents the "true" concentration?" *by* K. Neitola et al.

Anonymous Referee #3

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The authors describe findings from a flow experiment investigating H2SO4 measurements using different detection techniques, i.e. a denuder technique measuring total sulphate (MARGA) and nitrate CIMS and CI-API-TOF measuring the H2SO4 monomer. Gas-phase H2SO4 is provided by flushing a part of the carrier gas through a saturator with concentrated sulphuric acid. The main message of this paper is that there exists a large discrepancy of about two orders of magnitude for the total sulphate concentration by MARGA vs. H2SO4 monomer concentration measured by the mass specs analysing the same sample. Furthermore, the MARGA results confirm roughly the ex-

pected H2SO4 concentration derived from the vapour pressure. The authors are not able to explain the large discrepancy although a state-of-the-art measurement technique was available in the experiments (CI-APi-TOF). At the moment, the whole story sounds a bit mysterious for me. Here my comments:

- Nothing is said regarding the CI-APi-TOF results other than H2SO4 monomer concentrations! Are there any other clusters visible? What's the matter with H2SO4-base or H2SO4-organic compounds? How the authors checked the carrier gas impurities? How can carrier gas impurities influence or explain the observed disagreement of total sulphate and H2SO4 monomer concentration?

- In a single paragraph the MARGA technique should be compared with CIMS and CI-APi-TOF stating the detection limits, upper detectible concentrations, calibration results and the expected measurement errors.

- The authors should explain more in detail equation (1) regarding the "simple" equation of H2SO4 vapour pressure as derived from experimental data by Ayers et al.! I guess it is not enough to reference only to the work by Kulmala and Laaksonen.

- The authors observed nucleation for H2SO4 concentrations of about 10(7) cm(-3) in line with former results from the same experiment but using another H2SO4 source. Do they believe that there was no third body involved in the nucleation process? They should explain their findings in light of the current knowledge of atmospheric nucleation.

- The work by Ball et al.(1999) and Zollner et al.(2012) has been mentioned in the manuscript. Both studies used also a liquid H2SO4 source and H2SO4 monomer measurements by CIMS. A discussion of the findings of this manuscript regarding the former papers would be fine.

Finally, I guess, the authors should try to find out the reason for the missing H2SO4. They have the best technique at the moment doing that.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 2313, 2013.

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Interactive Comment