Atmos. Chem. Phys. Discuss., 10, C11885–C11887, 2011
www.atmos-chem-phys-discuss.net/10/C11885/2011/
© Author(s) 2011. This work is distributed under
the Creative Commons Attribute 3.0 License.

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Homogenous nucleation of sulfuric acid and water at atmospherically relevant conditions" *by* D. Brus et al.

Anonymous Referee #1

Received and published: 3 January 2011

The authors present measured nucleation rates of the system sulphuric acid/water as a result of flow-tube experiments and show a comparison with field measurements. Data for J are given as a function of RH for constant temperature as well as for nearly constant RH as a function of temperature. All particle measurements have been done using a state-of-the-art PSM counter with a cut-off-size of 1.5 nm and a modified commercial TSI3025 UCPC allowing the detection of particle down to a mobility diameter of 2.28 nm. Usage of a 1.5 nm counter allows observation of freshly produced particles close to the critical cluster size. This manuscript is within the scope of this journal. The atmospheric process of new particle formation is still a hot topic in atmospheric science. Although there is a lot of progress in the last time, not all questions are resolved jet. So the experimental findings of this work could help for a better understanding of the processes going on in sulphuric acid/water nucleation. Some minor critical points

could be considered before publication:

1) I guess, this is the first experimental work describing temperature-dependent measurements of J using a high efficient particle counter with a cut-off-size of 1.5 nm. This fact should be highlighted also in the Abstract. Moreover, a discussion is missing regarding the T-dependence of sulphuric acid/water nucleation generally and a comparison with other literature data published so far.

2) It should be stated what kind of changes were done to improve the 50% cut-off-size of TSI3025 from normally 3 nm to 2.28 nm. In tables and figure captions there should be a note pointing to the improved counting efficiency.

3) Nucleation rates observed by means of PSM are higher than those from TSI3025 measurements. This fact is more pronounced for relatively low sulphuric acid concentrations and is qualitatively in line with measurements given by Sipilä et al., Science, 2010. Can the observed differences of both counters be explained by particle growth from 1.5 nm to 2.28 nm governed by sulphuric acid or are additional substances needed?

4) For ammonia an upper limit of 0.5 ppb is stated. Can the author give a rough estimated regarding the background concentrations of amines in the carrier gas?

5) The given slope from PSM measurements at 25C and RH=57% in table 3 is 1.7. In table 4 a slope of 1.2 is given for 25C and RH ca.50%. More information is needed with respect to the precision/repeatability of the experiments. The error limits of deduced parameters (slopes, k-values) are helpful. What are the consequences for the trends measured by varying RH or temperature?

6) Sulphuric acid measured by CI-MS accounts for only 20% (RH=50%) or 1% (RH=16%) of the theoretical value expected from sulphate balance. Insufficient CI-MS detection efficiency of bound sulphuric acid is discussed. In the former paper of the FMI experiment, Brus et al., ACP, 2010, there were also shown a strong disagree-

ments between measured sulphuric acid (bubbler with subsequent IC analysis instead of CI-MS!) and the sulphate balance. Here, a more detailed discussion of the experimental findings with both approaches is necessarily needed. Nevertheless, the proposed “shielding effects” are very interesting also with respect to atmospheric sulphuric acid measurements!

7) There are some typos, e.g. notation of nitrate ions, etc.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 25959, 2010.