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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ternary homogeneous nucleation of H_2SO_4, NH_3, and H_2O under conditions relevant to the lower troposphere" *by* D. Benson et al.

Anonymous Referee #3

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The formation of new particles in the atmosphere is still a hot topic in atmospheric science. Although in the last time there is a lot of progress open questions remain.

The authors show experimental findings from a flow-tube experiment investigating the role of NH_3 additions for H_2SO_4/H_2O nucleation. H_2SO_4/H_2O is formed via the reaction of OH with SO_2. Experimental conditions are close to atmosphere, the residence time in the tube is in the range 60 – 240 sec. H_2SO_4 and NH_3 concentrations are obtained by means of a mass spectrometer. This manuscript is one of a series of papers by this group. A paper with similar topic was already published last year, Benson *et al.*, GRL, 2009.

1. I am very confused by comparing the data given in this manuscript with earlier results from the same group using more or less the same experimental approach. The only change in the setup seems to be the enlargement of the tube diameter from 2.54 or 5.08 cm to 13 cm at present. With the smaller tubes a H_2SO_4 concentration of 10^8-10^9 molecule cm^{-3} was reported for a nucleation rate of unity (Young *et al.*, ACP, 2008). But in this manuscript data are shown for J = 1 $cm^{-3}s^{-1}$ for a few of 10^6 molecule cm^{-3} of H_2SO_4 (also in the absence of NH_3 addition). Nothing is given in the manuscript explaining this discrepancy! Lowering of the importance of wall losses with the new tube cannot be the reason for this large difference, in the former studies the H_2SO_4 wall loss was intensively discussed by the authors.

2. The authors used for particle monitoring a TSI 3786 counter and they are stating that particles with a diameter $>$ 3nm were detected. Assuming a critical cluster size of 1-1.5 nm a growth of about 1.5-2nm in diameter is needed. This cannot be explained by 10^6-10^7 molecule cm^{-3} of H_2SO_4! A clear statement regarding the growth processes in the tube incl. the H_2SO_4 limited growth is needed.

3. In recently published papers (Sipilä *et al.*, Science, 2010; Berndt *et al.*, ACP, 2010; Brus *et al.*, ACPD, 2010) the importance of high efficiency counters with a cut-size of about 1.5 nm for nucleation experiments with low growth times was clearly shown. Application of less efficient counting devices should result in an underestimation of total particle numbers and in an overestimation of the slope log(J) vs. log (H_2SO_4). The authors are stating slopes of 3.6-4.6 being definitely higher than those given in the three papers above. On the other hand, the given values for J are close to (or somewhat higher than) the observation from the other experiments in literature. Also this topic should be discussed in this paper.

4. Was NH_3 measured only at the entrance or also at the tube outlet? What was the NH_3 loss in the tube?

5. It is stated that NH_3 was flushed into the tube together with the water vapour resulting in NH_3 mixing ratios of 20-100 pptv. What are the background concentrations of amines and organics in the flow tube? Especially in the case of amines, very low concentrations of these substances can clearly influence nucleation.

From my point of view a main revision is needed.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 22395, 2010.