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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Experimental investigation of homogeneous freezing of sulphuric acid particles in the aerosol chamber AIDA" *by* O.Möhler et al.

Anonymous Referee #2

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General points:

This is a very interesting article on homogeneous ice nucleation in H2SO4 aerosols. The laboratory study presented here is probably the one that comes closest to processes in the real atmosphere that has published so far on this topic.

I think the conclusions are significant, and that the length and detail are appropriate for publication in ACP. I recommend publication after the following main points and a number of minor/technical points have been taken into account.

Major scientific points:

p.1447, l.27–...: The two experiments at T=218K and $S_{nuc} = 1.29$ are considered to be outliers. I am not sure that this is correct. Looking at all the points of series A they all fall on a line that has a steeper slope than the experiments of series B and C. It appears that the whole series A is systematically different from the series B an C. This is further corroborated by comparing the H2SO4 concentrations of the aerosols in series A in the experiment at 218 K (A6_1 and A6_2) with the ones at 190 K (A1_1, A1_2, and A2_1): the droplets freezing at higher temperatures had a larger concentration than the ones freezing at lower temperatures, which is unreasonable, unless something else was happening. These inconsistencies are not present in series B and C, so I am really wondering what was going on in series A. From what I see, series A was the only one relying on the ACMS to determine aerosol composition and aerosol water content. Is there any chance that this might have influenced the results? If not, what else could be the reason? I don't think that the results can be explained by heterogeneous ice nucleation on incompletely removed background aerosol particles. According to Table 1, the experiments at 218 K were the last in the series (A6_1 and A6_2), so it is unreasonable to see that all the experiments before were not affected. Wouldn't one expect the heterogeneous ice nuclei to be removed by sedimentation by the time the A6_1 and A6_2 experiments were performed?

On p.1436 l.5–10: Can temperature fluctuations be responsible for the different response in series A? Such temperature fluctuations would give results as observed in A6_1 and A6_2: lower mean saturation ratios than needed for homogeneous ice nucleation to occur (only the negative eddies in T do reach the required supersaturations). Is there any evidence that T fluctuations might have been stronger in the A6_1 and A6_2 experiments?

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Interactive Comment

Minor and technical comments that should be considered in the revised version:

(1) p.1430, l.21: 'Instead of Freezing nucleation in...' put 'Homogeneous freezing nucleation in...'

(2) p.1431, l.13–16: You should make a distinction here between ice and nitric acid hydrates. While there is a consensus in the case of ice, I believe there exist different opinions whether or not nitric acid hydrates freeze homogeneously from STS.

(3) p.1432, l.20: Replace 'super-cooled' with 'supercooled'.

(4) p.1432, l.24: Does this have implications with respect to the applicability of the present study to atmospheric conditions? If not, please state so, if yes, explain.

(5) p.1433, l.16: Replace 'are evaporated' with 'is evaporated'.

(6) p.1440, l.5: What exactly do you mean by 'ex situ'? Just that the FISH instrument was operated outside of the chamber?

(7) p.1440, l. 12: The detection limit refers to a mixing ratio. At what pressure? Or better, what is the detection limit in terns of H2O partial pressure?

(8) p.1443, l. 9–15: You speculate only about the first very sharp increase of the signal, not about the second rise, which I think is the more peculiar one. What happens at that sharp rise in signal about 50 s after t_{nuc}?

(9) p.1444, l.2: Replace 'Maximal' with 'Maximum'.

(10) p.1445, l.19–23: I understand that ice particles need to grow beyond a critical size before they can be detected as individual particles in the ACMS. However, before that occurs one would expect an increase in the water content of the continuous background particle signal. I cannot see such an increase in Figure 6f. What is the reason?

(11) p.1445, l.23: Please add the ACMS uncertainty of 30 s to the last column in Table

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2 in order to have the uncertainties for all instruments in the table.

(12) p.1447, l.15: Replace ‘contributions’ with ‘contribution’

(13) p.1447, l.15: The formula looks strange. I suppose you mean $\ln S$ not $l\,n\,S$, don’t you?

(14) p.1450, l.6, 13, and 19: ‘supercooled’ instead of ‘super cooled’ and ‘supersaturated’ instead of ‘super saturated’

(15) p1456, Table1: To the general reader it would be more helpful to see the cooling rates $(dT/dt)_{nuc}$ or the change in saturation ratio $(S_{ice}/dt)_{nuc}$ displayed for each experiment rather than $(dP/dt)_{nuc}$. Please add these to the table.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1429, 2002.