

Reviewer Comments for Manuscript acp-2010-884: *New cloud chamber experiments on the heterogeneous ice nucleation ability of oxalic acid in the immersion mode.*

General Remarks:

The above article presents and discusses ice nucleation behaviour of oxalic acid (OA) in ternary solutions of both OA/NaCl/H₂O and OA/H₂SO₄/H₂O. In addition experiments are presented with discussions on the ice nucleation of NaCl and H₂SO₄ solutions. A detailed comparison is presented to the study of Zobrist *et al.* [2006]. The results are of interest to the readers of Atmospheric Chemistry and Physics. The results *could* have implications for mixed-phase cloud formation.

I recommend the manuscript for publication after some adjustments have been taken into account.

In its current form the manuscript is drafted in a manner that many at times suggests that the goal of the study was to compare and contrast their work to that of Zobrist *et al.* [2006]. While this is a useful comparison, it should not appear like this is the goal of the paper.

The manuscript is also quite long. I understand the reason for this given that all details of the experimental set up, results and discussions are outlined very well and comparisons have been performed in detail. However to make the manuscript more reader friendly I suggest splitting the paper into more sections/sub-sections. There is plenty of room in the manuscript for further organisation/division.

For example, on page 29454, there is a substantial amount of discussion about the experimental set up and procedure of the conducted experiments. That should be put under the experimental section maybe under a subsection of “experimental preparation” or something of the sort. Alternatively it could also come at the end of the introduction section. On this page (29454) the authors also describe the goals and objectives of the current study leading readers to believe that the introduction is coming to an end. However, the introduction continues (and rightfully so) for 4 more pages at which stage more objectives of the current work are mentioned. I suggest these two sections of objectives should be merged.

In terms of forming more subsections in the paper, perhaps the authors could maintain the approach used. i.e. two results sections, one for NaCl/OA and the other for H₂SO₄/OA. However, under each section the results could be split between comparisons of first expansion cycles and then another section for comparisons between second expansion cycles since all the ice nucleation figures are presented as such.

Also where ratios have been expressed they would be better visualised by the reader if the format 1:XX was used by the authors rather than using for example, 5:0.51. The 1:XX format allows for a quicker understanding of the relative contribution of oxalic acid in each of the solutions used.

Specific Remarks:

Pg. 29450

Line 13: delete the word 'on'

Line 14: replace 'again' with 'subsequently'

Pg. 29453

Line 12-14 should read as follows:

'....Zobrist et al. (2006) detected that homogeneous ice nucleation leads to the nucleation of crystalline organics in aqueous solution droplets. After having...'

Pg. 29454

See comment above in 'General Remarks' section.

Pg. 29455

Line 8: replace 'that' with 'the procedure' and insert a comma after '235 K'

Pg. 29456

Line 1: Delete 'Already in the introductory part of our manuscript' and start the sentence with 'We..'

Pg. 29460

Line 11: delete 'so' from 'so-induced' and insert a comma after 'rate'

Pg. 29461

Line 12: insert the word 'and' after 'detected'

Line 28: replace manuscript with 'work'

Pg. 29462

Line 17: replace 'the so-' with 'such'

General question: What is a two-stream dispersion nozzle? Does this mean that the cloud chamber is fed from two different inlets that have been split from the same source of particles?

Pg. 29463

Line 20: Insert the AIM thermodynamics model II reference here

Pg. 29465

Line 10: replace 'this manuscript' with 'section XX' (the idea is to be specific in pointing readers where to look for information. This will also help once the paper is organised into more sections)

Line 23: what is meant here by '....suitable sites in the chamber interior'? Explain here if you mean background aerosol? Is there a high enough background to instigate crystallisation of 10% of the particles?

Line 29: replace 'is' with 'are'

Pg. 29468

Line 4: ‘aerosol loading’ should be changed to ‘aerosol load’ and ‘whose records are’ should be replaced with ‘as’

Line 16: delete ‘so-’ and ‘at’ and insert ‘where’ before ‘their’

Line 26: the word ‘section’ is missing after the word ‘following’

Pg. 29476

Line 28: The authors mention a reduction in the $S_{ice,crit}$ from 1.38 to 1.31 and 1.33. How significant is this reduction. It should be presented within the context of how accurate or what the uncertainty of measurement is in AIDA RH_{ice} . In the past some AIDA measurements [Möhler *et al.*, 2005] have reported uncertainties in water vapour concentrations of ± 5 to $\pm 10\%$. This could translate into an uncertainty higher than $\pm 5\%$ in S_{ice} , therefore influencing the significance of some of the results. This aspect should be clarified.

Pg. 29481

Line 4: replace ‘loading’ with ‘load’

Pg. 29482

Line 8: replace ‘(results not shown explicitly)’ with ‘(data no shown)’

Pg. 29483

Line 3: at the end of the line ‘estimate’ should read ‘estimates’

Line 6: ‘...in the AIDA chamber...’

Line 29: replace ‘Afterwards’ with ‘we then changed’

Pg. 29484

Line 1: delete ‘was changed’

Line 5: delete ‘have’

Pg. 29486

Line 8: delete ‘so-’

Pg. 29487

Line 4: replace ‘happened’ with ‘occurred’

Line 24: insert a comma after ‘ratio’ and after ‘ $S_{ice,crit}$ ’

Pg. 29488

Line 6: The authors state here that their findings corroborate the assumptions that oxalic acid might play a considerable role in the Earth’s climate system. I think this is a little far fetched, especially since they state that it is based on an assumption. Given the small fraction of oxalic acid particles that acted as heterogeneous IN, one could argue that these particles may contribute to ice formation in the atmosphere, but is their contribution to ice in mixed-phase clouds nearly as much as that of mineral dusts for example? The findings from the current paper are of importance to gaining mechanistic knowledge of the behaviour of oxalic acid, or even low-solubility di-carboxylic acids within the

framework of freezing and crystallisation in atmospherically relevant aerosols, however, statements about ‘playing a considerable role in the Earth’s climate system’ can only be made after evidence from a model run for instance. Definitely in a GCM the current results would not show any influence on ‘climate forcing’, however, it is possible the results are implemented in small scale and regional models, one may see an influence due to ice heterogeneous ice nucleation of di-carboxylic acids.

Line 11: replace ‘indispensable’ with ‘important’

Figures and Tables

Where ratios have been used, consider switching to a more standard format of 1:XX (see comment above in general remarks section)

Table 2, caption:

Line 1: replace ‘about’ with ‘of’

Line 7: replace ‘of’ with ‘for’

Figure 1, caption:

Line 4: ‘...shown as THE black line,...’

Figure 5, caption;

The font appears to be smaller than the rest of the figures.

Figure 6, caption;

Line 1: Insert comma after ‘...ratio’ and before ‘from’

References

- Möhler, O., S. Buttner, C. Linke, M. Schnaiter, H. Saathoff, O. Stetzer, R. Wagner, M. Kramer, A. Mangold, V. Ebert and U. Schurath (2005). "Effect of sulphuric acid coating on heterogeneous ice nucleation by soot aerosol particles." *Journal of Geophysical Research-Atmospheres* **110**(D11): doi:10.1029/2004jd005169.
- Zobrist, B., C. Marcolli, T. Koop, B. P. Luo, D. M. Murphy, U. Lohmann, A. A. Zardini, U. K. Krieger, T. Corti, D. J. Cziczo, S. Fueglistaler, P. K. Hudson, D. S. Thomson and T. Peter (2006). "Oxalic acid as a heterogeneous ice nucleus in the upper troposphere and its indirect aerosol effect." *Atmospheric Chemistry and Physics* **6**: 3115-3129.