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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "New cloud chamber experiments on the heterogeneous ice nucleation ability of oxalic acid in the immersion mode" *by* R. Wagner et al.

R. Wagner et al.

robert.wagner2@kit.edu

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We thank Referee #4 for the careful manuscript reading. Below we will address the individual comments.

COMMENT: I recommend to publish the work in ACP after carefully editing. The discussion of all details on all associated measurements is one of the causes that the overall length of the paper exceeds the standards of this journal. Beside this, as a reader I tent to "get lost" in the explanations of the many aspects. Which means - the main message is hard to follow. Furthermore the authors tent to mix sections. For example there is a large subsection in the "Introduction" (P 29454, l.18pp) where AIDA experimental details are discussed which are partly repeated in the following sections (where they are supposed to be). Also in the following introduction section (P 19456 l.1pp) the differences between AIDA and Zorbrist et al.2006 measurement are discussed in detail and in section 3 again (P29485 L27 pp). The challenge is now to edit this paper by structuring and distilling the main messages. Here it would help to devide the quite long sections in subsections with individual captions which will guide the reader more gentle.

ANSWER:

As outlined in detail in our response to Referee #3, we have made improvements regarding the manuscript organisation, including new subsections and new introductory comments before each main section. Specifically addressing the concerns of Referee #4, we have split the introduction into two parts, emphasizing that the new section 2 ("Experimental strategies") describes the general differences in the experimental approaches between the cloud chamber and emulsion freezing experiments whereas the new section 3 ("Methods") addresses the details of the technical operation of the AIDA chamber. We consider it necessary to re-address in our old section 3 (P29485 L27 pp) the differences between the cloud chamber and emulsion experiments because we here propose an explanation to bring our and the Zobrist et al. (2006) findings into agreement. To better guide the reader, we made a new subsection out of this paragraph (section 4.2.3 "Conclusions", see answer to Referee #3).

COMMENT:

Minor details: The thesis of the potential importance of particulate oxalic acid has to be checked by discussing the relative role of the ice nucleation ability in comparison to e.g. mineral dust.

ANSWER

As also outlined in detail in our response to Referee #3, we have re-arranged and

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revised our summary section, now including a paragraph where we compare the heterogeneous ice nucleation ability of oxalic acid dihydrate in the immersion mode with recent findings for other atmospherically relevant aerosol species like mineral dust and soot.

COMMENT:

In "AIDA-Plots" Fig. {5; 7; 10; . . . ; 17} I would like to see in the scatter plots (particle size vs. time) the median, 25, 75 percentile of the particle size in addition to the scattered single data- even if the complex diagram becomes more complex. But this data will make the "dot cloud" more quantitative readable.

ANSWER:

We somewhat refrain from following this suggestion because, as emphasized in the manuscript, for aspherical particles (e.g. ice crystals, effloresced NaCl crystals) the size classification by the OPCs is ambiguous. In contrast to spherical particles, it depends on the accidental orientation and the shape of the aspherical particles. The only quantitative information from the OPC data that is used in our analysis is the ice particle number concentration and the calculated ice-active fraction. These results are directly plotted below the scatter data in each graph. We additionally show the scatter data because – in our opinion – they are an illustrative tool to guide the reader through the complex AIDA expansion experiments. It becomes easier to follow the different periods of an expansion experiment (e.g. in the left part of Fig. 5: deliquescence of NaCl, formation of a droplet cloud, and finally, ice nucleation).

COMMENT:

(P29461 l. 15) The definition of the depolarization ratio should be: delta = (I_per – I_par)/(I_per + I_par) . . . to avoid a potential singularity if the scattered light is completely perpendicular polarized.

ANSWER:

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The most general meaning of the linear depolarisation ratio as defined by I_per/I_par is the relative change of the linear polarisation state of the incident light due to the single scattering process. This includes the formation of partly non-polarised light and the rotation of the polarisation vector. In the case of ice crystals, a rotation of the incident polarisation vector by 90°, which is necessary for a potential singularity, would require perfectly oriented particles for the formation of specific spatial skew rays which rotate the incident vector by internal reflections (Takano & Jayaweera, 1985). In our experiments, the ice particles are randomly oriented so that a singularity can be excluded. In the case of small (aerosol) particles, maximum depolarization ratios of 0.7 have been calculated by Mishchenko & Sassen (1998) for randomly oriented spheroidal and cylindrical particles. Moreover, the definition given above is widely used for the representation of atmospheric polarisation lidar measurements.

Mishchenko, M. I., and Sassen, K., Depolarization of lidar returns by small ice crystals: An application to contrails, Geophys. Res. Lett., 25, 309-312, 1998.

Takano, Y., and Jayaweera, K., Scattering Phase Matrix for Hexagonal Ice Crystals Computed from Ray Optics, Appl. Opt., 24, 3254-3263, 1985.

COMMENT:

What is the effect of multiple scattering on the depolarization ratio if the cloud becomes thick (up to 80 $cm{-}3$)?

ANSWER:

The effect of multiple scattering on the linear depolarisation ratio was investigated e.g. by Ryan et al. (1979). They found that the backscattering linear depolarisation ratio becomes significantly influenced by the multiple scattering only when the extinction coefficient of the cloud exceeds a value of about 0.1 per meter. The highest extinction values during the AIDA expansion experiments are not encountered for ice clouds but are observed for dense droplet clouds, e.g. during the time period spanned by the two

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vertical green lines in the left panel of Fig. 5 from Exp. 1. Even when using a mean droplet size of 10 micrometer (which is in fact the upper droplet diameter observed in the experiments) and a number concentration of 500 per cc, the extinction coefficient is only about 0.05 per meter which could increase the depolarization ratio of the droplet cloud by just a few percent.

Ryan, J. S., Pal, S. R., and Carswell, A. I., Laser Backscattering from Dense Water-Droplet Clouds, J. Opt. Soc. Am., 69, 60-67, 1979.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 29449, 2010.

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