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Atmospheric Chemistry and Physics Discussions

Interactive comment on "In situ observations of aerosol particles remaining from evaporated cirrus crystals: Comparing clean and polluted air masses" *by* M. Seifert et al.

B. Kärcher (Referee)

bernd.kaercher@dlr.de

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The paper describes in situ measurements of aerosol particles contained in cirrus ice crystals. The authors employ advanced methods to measure and analyze interstitial and residual particle size distributions. The focus is to work out interhemispheric differences between the properties of these particles. The underlying measurements have been performed at midlatitudes during the INCA project.

My overall impression is that this paper represents an important contribution to current research on cirrus clouds. It will be a strong publication in ACP after consideration of the points listed below. Most of my comments concern the Discussion in section 4.

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Detailed comments

p.1601, l.2-7
The authors suggest that a very weak aerosol-cirrus link exists in general. However, the cited papers by Jensen and Toon and Kärcher and Lohmann are not making such a point. It is only for liquid supercooled aerosol particles undergoing homogeneous freezing that the number of ice particles is rather insensitive to variations of aerosol size and number.

I suggest to delete the passage ", whereas the nature .. a minor role" and the beginning of the next sentence "On the other hand", that should now start with "Very little is actually known ...". The whole paragraph may then be merged with the next one starting on line 8.

p.1601, l.10
Importance of the small (how small, by the way ?) aerosol particles for what ?

p.1609, l.1-4
You may wish to say something about the similarity between the size distribution of the residuals and the size distribution of the aerosol particles that actually have formed ice.

In particular, consider the two following mechanims: (i) if fractal particles freeze, the geometric structure of those particles might change once they evaporate all water molecules. The "size" of the fractal particle before and after cloud formation may be significantly different. The change in aerodynamic properties due to agglomerate collapse is a well known effect (e.g., refer to Ramachandran and Reist, Aerosol Sci. Technol., 23, pp.431, 1995; Leong, J. Aerosol Sci., 12, pp.417, 1981). (ii) Scavenging of interstitial particles during the cloud lifetime and sticking together of the scavenged particles after evaporation could lead to fewer small particles in the residual spectrum as compared to the spectrum at the point of freezing. The efficiency of this mechanism will depend on cloud surface area and cloud lifetime, and may be relevant in long-lived, extended cirrus.

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p.1614, l.17
With ".. the temperature asscociated with homogeneous freezing .." you probably refer to the spontaneous freezing limit of pure water drops. In fact, spontaneous freezing in supercooled water occurs somewhere between 235 and 240 K; the exact temperature depends on the freezing rate and on the definition of freezing onset and is difficult to measure.

It is not clear if you refer to the above spontaneous freezing limit, or to freezing temperatures of supercooled liquid aerosol particles. The latter depend on water activity, too, the upper bound being the spontaneous freezing of pure water drops. Please indicate that you actually refer to the latter.

p.1614, l.23 – p.1615, l.2
It is stated that there is "no significant statistical difference" in residual properties in different temperature ranges and you find that surprising.

To my understanding, in the region 238-243 K, aerosols are activated into water droplets prior to freezing. Below 235 K, cirrus ice particles form from freezing of aerosol particles. In between, both freezing paths may take place. Now, the freezing conditions in these 3 regions may differ from each other; the mode of nucleation may change; and the vertical velocities in the selected temperature regimes may not be the same. These differences might actually mask possible differences to be expected compared to the cases when only temperature is changing, especially if these differences occur in a random fashion.

Please either clarify this or remove this paragraph if I could convince you that your finding may not be surprising.

p.1618, l.6-8
Please check in the light of the above comment (p.1614, l.23 – p.1615, l.2). Also, the mean temperatures and temperature distributions were relatively similar in both campaigns. Together, this probably explains the relatively little variance of residual

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measurements with temperature.

p.1618, l.9
This again refers to the last two remarks. I am not sure if you can really state that this is unexpected on the basis of homogeneous freezing. All the effects you mention in this para do not necessarily imply that residual distributions should strongly depend on temperature in the range between 235 and 243 K.

p.1618, l.25-26
If you drive homogeneous nucleation with a wide range of vertical winds, you will get a wide range of ice crystal concentrations, too. I do not deny the potential importance of heterogeneous processes, but advise you to make a more careful statement.

Your statement about the potential importance of heterogeneous ice nuclei is supported by the observations shown in Figure 8, and in the discussion this point should be made more clearly. Whether heterogeneous processes control the wide variance of observed ice crystal concentrations requires further analysis.

p.1619, l.9
The simulations you cite do not really suggest that heterogeneous nucleation occurs at lower relative humidities, the lower freezing thresholds are more or less prescribed. It may well occur that the difference in freezing threshold relative humidities between homogeneous and heterogeneous freezing modes is not significant, if insoluble particles are no potent ice nuclei.

p.1621, l.8-9
You should at least add: "... within the temperature range 235-243 K."

p.1633, caption Figure 8
The text written on p.1616, l.14/15, should be noted in this figure caption (fraction is only estimated below $D = 0.025\mu$m.

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Typos

p.1601, l.11
"analyses" instead of "analysis"

p.1603, l.4
"calculate" instead of "calculates"

p.1614, l.17 and p.1623, l.14
"homogeneous" instead of "homogenous"

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1599, 2002.