Atmos. Chem. Phys. Discuss., 3, S1983–S1990, 2003
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol-cirrus interactions: A number based phenomenon at all?" *by* M. Seifert et al.

M. Seifert et al.

Received and published: 21 November 2003

Major comments:

reviewer: "This paper relates the number concentration of ice crystals Ncvi to the number concentration of interstitial aerosol Nint. By addressing the similarities/ discrepancies to the relationship of aerosol number concentration and cloud droplet number concentration, the authors are able to speculate on the physical mechanisms causing the positive and negative correlations. While the positive correlation between Nint and Ncvi is similar to what is found in warm clouds, there is no analogous situation for a negative correlation in warm clouds. Here the authors speculate on 2 different hypotheses for a sublimating cloud, suggesting that either pollution retards the sublimation of cirrus clouds or that the sublimation of cirrus clouds leads to new aerosol particle formation. Since this unexpected anticorrelation is a central part of this paper, it would be nice if the authors could try to substantiate each hypothesis. In particular, if the sub-

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limation of cirrus clouds is a source of aerosol particles, how are you suggesting that this happens? Are you suggesting that aerosols form by nucleation in the vicinity of the cloud as discussed by Perry and Hobbs (1994) for warm clouds? If so, I would argue that you do not have the right instrument to prove that because these aerosols should be smaller. Also since aerosol nucleation depends on temperature, aerosol precursors and aerosol surface area, you should evaluate the temperature, aerosol chemical composition and aerosol surface area as a function of Nint and Ncvi. Or are you thinking along the lines of ice crystals leaving behind an aerosol residue? I suspect that this mechanism would not increase the number of aerosols sufficiently with what you have indicated with arrow 2 in figure 10. Here Nint increases from 50 to 300/cc while Ncvi decreases from 1/cc to below 0.01/cc. So, how can the sublimation of Ncvi explain the increase in Nint? I understand that it is beyond the scope of this study to evaluate the chemical aerosol composition in the entire data set but could you try to analyze smaller flight segments in more detail, like the one shown in figure 2 to prove or negate one pathway? As mentioned above, I would strongly encourage the authors to try to substantiate their different hypotheses better before publication of this manuscript."

reply: The ambition of this work was to study the aerosol-cloud interaction in much the same way as it has been done previously for warm clouds. Since this was the first attempt to do this for cirrus clouds we felt that there was a limit to how extensive the analysis could be on findings that came up as a result of this work. We see this work as a two step process where this study helps establishing observed relations and a follow up study(s) will have to address the findings more in depth.

At this stage we spent some effort to show that the relations we observe are not bogus and that we need to understand them better, just as the reviewer suggests. However, even if we limit ourselves to work with a case study, we believe that the additional material will have to be substantial in order to establish general relations. Since we are essentially left with speculation about the possible mechanisms we suggest, we intentionally kept them brief. The reasoning by the reviewer suggests that we were

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perhaps too brief. We therefore elaborate a little more on these issues based on the comments by the reviewer to make it a little more clear.

Action: rephrase the discussion section (p. 3642 line 27 to p. 3643 line 15): “Assuming that the amount of aerosol particles is a proxy for the degree of pollution, cirrus crystals may incorporate solutes onto the crystal which are produced by gas phase reactions or heterogeneously on the particle surface. When a crystal is relatively large this surface contamination may have little influence on the transfer of water vapor to or from the crystal as the relative humidity changes. As the crystal evaporates and becomes smaller the presence of this surface contamination, or impurity, may become important. The threshold could be reached at a level where one or a few monolayers are covering the evaporating crystal preventing water vapor to have full access to the ambient environment. In other words, surface effects may suppress evaporation rates which would be consisted with results by Chen and Crutzen (1994). Chen and Crutzen (1994) studied the possible influence of trace chemicals on the growth and lifetime of ice crystals, based on the concept of a transition layer on the crystal surface. They showed theoretically that ice crystals may survive without complete evaporation at relative humidities over ice as low as 91%.

The other interpretation refers to the possible production of particles. As pointed out above, ice crystals may scavenge chemical species from the interstitial air. When an ice crystal evaporates, the release of the scavenged species may result in locally enhanced concentrations which could favor aerosol formation. The evaporating cloud, as a source of particles, could explain the shift towards more negative correlations below ice saturation in Figure 5. If this particle production mechanism is real, cirrus clouds could constitute a very significant source of new particles in the tropopause region. Note that in the CVI crystals are transported in nitrogen gas covered from sunlight. Hence we donŠt expect particle production to be possible within the CVI from the same mechanism.”

references: Chen, J.P.. and Crutzen, J.: Solute effects on the evaporation of ice parti-

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cles, J. Geophys. Res., 99, 18847-18859, 1994

reviewer: “More serious and even harder to understand are the results displayed in figure 7 for RHi > 100%. Why should Ncvi decrease with increasing Nint? I could understand a leveling off or a saturation effect of Ncvi with increasing Nint but not a decrease in Ncvi. From my point of view, the most likely explanation is that the ice crystals are too small to be detected. However, if that is the case, then the ice crystal data shown here are due to a sampling artifact for a large range of Nint, which questions the usefulness of this large part of the data set and the discussion in the paper. Are there no other ice crystal measurements that could be used, such as the FSSP to address this problem?.”

reply: The reviewer make a very good point here. To address this issue we follow the suggestion by the reviewer and include FSSP size distribution data into the analysis. Using the crystal size distribution data we can derive the number density of particles smaller than the cut-off size of CVI (NFSSP-1). Plotting NFSSP-1 as a function of Nint as was done for the CVI data shows high crystal number densities at low aerosol number densities and low crystal number densities at high aerosol number densities. In other words the observed relation between Ncvi and Nint does not seem to be due to a sampling artifact due to the CVI cut-off size.

action: We will include the following paragraph starting at p. 3637 l.14 :

“When interpreting crystal residual data we have to keep in mind that the CVI only detects particles larger than ca. 5 μm in size, and even if the cloud is composed of a large number of crystals these will not be detected should the size be too small. Thus, there is a possibility that the crystal number density continues to increase with increasing Nint, but remains undetected by the CVI as the mean size is smaller than the detection limit of the CVI. To address this issue and to verify the in Figure 7a,b found relations between Ncvi and Nint we extend our analysis by including FSSP-300 crystal size distribution data. A lower limit of 4 μm for the FSSP size distribution has

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shown to give the best agreement between FSSP and CVI data. Integrating the FSSP size distribution from 4 to 15.8 μm (NFSSP-2) allows us to emulate a CVI with respect to the cut-off size. Recall that the CVI selects particles based on the aerodynamic properties of the crystal whereas the FSSP-300 inverts the light from a particle based on assumed optical properties of the crystal. To get a measure for the number density of small ice particles we integrate the FSSP size distribution from 1 to 4 μm (NFSSP-1). The lower integration limit was chosen to be above the lower detection limit of the FSSP to reduce the effect that large aerosol particles might have on the FSSP-300 data. In Figure 7c,d we plot NFSSP 1, NFSSP-2 together with Ncvi as a function of interstitial aerosol number density. When interpreting FSSP-data we have to keep in mind the FSSP data is biased towards low crystal number densities due to counting statistics. Nevertheless close agreement between NFSSP-2 and Ncvi data is found over the entire Nint range highlighting the consistency of FSSP and CVI measurements yet based on totally different physical principles. The NFSSP-1 data is shifted towards high number densities but resembles the same main features as found for NFSSP-2 and Ncvi; high crystal number density at low Nint and low crystal number density at high Nint. Hence the number of small ice crystals particles does not appear to increase with increasing aerosol number density, but rather mimics the trend for the larger crystals."

Minor comments:

reviewer: "p.3626, l.6: From (not form)"
reply: ok

reviewer: "p.3626, l.12: Related with (not to)"
reply: We do not agree and leave this unchanged

reviewer: "p.3626, l.24/25: Which mechanism is more likely?"
reply: At this point we are unable to say which of the mechanisms that would be most

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likely and more studies are required.Ť

reviewer: “p.3627, l.2: Change "the climate from influence on" to "on climate by influencing”
reply: ok

reviewer: “p.3629, title: Add "Data" after Experimental Please add the measurement uncertainty or error for each instrument.”
reply: ok

reviewer: “p.3631, l.6: remains (add s)”
reply: ok

reviewer: “p.3633, l.13: Delete: "above as well as colder colors below ice saturation”
reply: We do not agree since there are opposite gradients above and below 100% RHi

reviewer: “ p.3634, l.19: Change: "let" to "allowed”
reply: ok

reviewer: “p.3634, l.25: Delete: "a of" before about 100 s.”
reply: ok

reviewer: “p.3635, l.7: Add: "-" before 0.2”
reply: ok

reviewer: “p.3636, l.8: Change "and we would exclude" to "and therefore we exclude”

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reply: ok

reviewer: “’p.3638, l.3: I am not sure that I understand the second simulation. Are you weighting the random ice crystal number concentration by the total observed ice crystal number concentration? If so, please add "total". If not, please clarify.”
reply: We will add the word ŞtotalŤ.

reviewer: p.3638, l.16: distributions (add s)
reply: ok

reviewer: “p.3638, l.23: I would argue that the w variations are not completely random, but that w has a minimum around Nint=100-300/cc. Please interpret the implications of this minimum for Ncvi as a function of Nint in figure 7. Or maybe add a plot of Ncvi as a function of w.”
reply: We agree with the reviewer that the variations of the vertical wind displayed in figure 9 may not be completely random. Note that the INCA data set is quite extensive, but still finite. Nevertheless, there are no obvious trends that would explain the observed relations with local maxima in Ncvi and the correlation coefficient in both data sets as presented in figure 7. What regards the relation Ncvi and w we have already provided figure 3, where we plotted w as a function of Ncvi and RHi.

reviewer: Şp.3641, l. 14: I donŠt understand this argument. Why should a further increase in Nint quench ice nucleation and thus lead to a negative trend between Nint and Ncvi?Ť
reply: We have try to make this point more clear see below:
action: Rephrase the corresponding paragraph (p. 36411 l.6-20):
“Why is there an initial positive relation between Nint and Ncvi which then turns into

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a negative one? Since the role of aerosols in the formation of clouds is still an open issue we are left to speculate what impact changes of aerosol number density have on atmospheric ice particle number densities. It might be that the number of interstitial aerosol particles is a proxy for the number of particles suitable for ice crystal formation. At low Nint ice crystal formation might be limited by the number of suitable aerosol particles, causing a positive relation between Nint and Ncvi. However, this trend cannot last indefinitely as the water vapor supply in the upper troposphere is limited, which is why cirrus formation tends to be self-limiting. The higher the number of suitable ice nucleating aerosols, the more particles will compete for the available water causing a decrease of the peak saturation. This will in turn lead to a decrease of the crystal number density, since fewer particles will simply grow to become real ice crystals and will remain as haze drops or ice embryos. At some point (or Nint) we therefore reach a situation where no more ice crystals can be formed, marking the maximum crystal number density for a given dynamical forcing. A further increase in Nint will quench ice nucleation increasingly, resulting in a negative trend between Nint and Ncvi. Note that a negative relation between crystal number density and interstitial aerosol number density at high Nint was observed for both CVI and FSSP data. Hence the possibility that the crystal number density may continue to increase with increasing Nint, but remains undetected by the CVI as the mean size is smaller than the detection limit of the CVI, can be dismissed."

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3625, 2003.