

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.

M. Seifert et al.

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First of all, we would like to thank Bernd Kärcher for their general and specific comments. The revised manuscript will incorporate almost all of the suggestions.

Here we address the specific points and questions of the reviewer.

1. p.1601, l.2-7 introduction

Reply: OK

2. p.1601, l.9-10 "importance of the small aerosol particles"

Reply: p.1601, l.9-10 will be changed to: "Noone et al. (1993) and Ström et al. (1997) presented the first size distributions of residual particles indicating the importance of

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submicron particles for cirrus formation."

3. p. 1601, l.1-4 *similarity between the size distribution of the residuals and the size distribution of the aerosol that actually have formed ice*

Reply: p.1618, l.1 the following text will be appended:

"One may speculate that the presented size distributions of residuals are greatly different from those of the aerosol particles that actually have formed ice. Ramachandran and Reist (1995) have shown that fractal particles become more regular when they are subject to condensation followed by evaporation. When an aerosol composed of irregularly shaped agglomerates is incorporated into an ice crystal, the agglomerates are likely to undergo morphological changes once the crystal evaporates. The size of a particle after a condensation and evaporation cycle may therefore be significantly different. Aerosol particles are likely to undergo repeated cycles of condensation and evaporation and any collapse of fractal geometries is only expected for the first cycle. Furthermore we have to keep in mind that fractal particles, which are most likely of carbonaceous origin, only account for a subfraction of the total aerosol population in the upper troposphere. For these reasons the process of condensation and evaporation is not likely to significantly change the size distribution of the particles that formed ice crystals.

Another process that might modify the residual particles is scavenging of aerosol particles from the interstitial air. Once the ice crystals evaporate in the CVI scavenged interstitial particles may stick to the particle on which the crystal originated. In other words, the crystals interact with the interstitial aerosol that in turn influences the residual particles by making them larger. We may try to estimate the result of this potential process by assuming an ice crystal lifetime of 3 hours, a crystal diameter D_{p1} of 10 μm , and an interstitial diameter D_{p2} of 0.03 μm . The interstitial number density N_2 is 100 cm^{-3} . The rate at which interstitial particles are transferred to one crystal is given by:

$$dN/dt = K_{12}N_2(s^{-1})$$

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The coagulation coefficient is defined as:

$$K_{12} = 2\pi(D_{p1} + D_{p2})(D_1 + D_2)(s^{-1}cm^3)$$

where D_1 and D_2 refer to the diffusion coefficients of the crystals and the aerosol, respectively. Whether scavenging affects the distribution depends very much on the size of the residual particles. Adjacent DMA bins differ in particle size with approximately 20%. Given the conditions above, only residual particles smaller than about $0.03 \mu\text{m}$ have a chance to grow that much. Thus we expect only a small or negligible effect on the smallest residual particles observed."

References:

Ramachandran, G. and Reist, P.C.: Characterization of morphological changes in agglomerates subject to condensation and evaporation using multiple fractal dimensions, *J. Aerosol. Sci.*, 23, 431–442, 1995.

4–7. temperature changes and the residual size distributions

Reply: p.1614, l.15 - p.1615, l.2 will be rephrased:

"We know that temperature is very important for cirrus formation and evolution. It is of special interest to investigate the residual aerosol distributions that are present near the temperature associated with homogeneous freezing of supercooled liquid aerosol particles (around 235 K). Each residual integral range contains size distribution measurements made at various temperatures, causing a mixing of data from different altitude levels. As we include a new parameter in our analysis we would like to stratify our data set further based on temperature. Ideally, we would like to keep the narrow $N_{CVI,10}$ binning and compare cases for a given residual number density and temperature. However, this is not practical because this would result in insufficient amount of data in most bins. For this reason we proceed by grouping the data according to temperature in three regimes warm ($238 \text{ K} < T < 243 \text{ K}$), intermediate ($235 \text{ K} < T < 238 \text{ K}$) and cold ($T < 235 \text{ K}$), and each regime is then divided into four residual number

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density ranges very high ($N_{CVI,10}$ 3-10 cm^{-3}), high ($N_{CVI,10}$ 1-3 cm^{-3}), intermediate ($N_{CVI,10}$ 0.3 – 1 cm^{-3}) and low ($N_{CVI,10}$ 0.1-0.3 cm^{-3}). Now, the freezing pathways are likely to be different in the three regimes. In the warm regime aerosols are activated into water droplets prior to freezing. Below $T < 235$ K aerosol particles freeze before water saturation is reached. For intermediate temperatures both freezing paths take place. In addition the mode of nucleation may change between the three temperature regions. In the warm and intermediate temperature regime both heterogeneous and homogeneous freezing processes may occur, whereas for $T < 235$ K homogeneous freezing is believed to dominate ice nucleation. Assuming that only the homogeneous freezing mode is active during ice nucleation, the freezing probability is a strong function size. For a given aerosol composition, solution droplets will nucleate starting from the large particle end of the aerosol size distribution. This results into a residual distribution where the number of residuals is dominated by accumulation mode particles, as observed for example for warm clouds (e.g. Glantz and Noone, 2002). However, no significant difference could be found between the size distributions based on different temperature intervals (Figure not provided). In fact comparing distributions between the different temperature ranges shows no significant statistical difference. This is a somewhat surprising finding given the expected importance of temperature in determining the mode of nucleation."

Reference: Glantz, P. and Noone, K.J.: Scavenging efficiencies of aerosol particles in marine stratocumulus and cumulus clouds. Submitted to Q.J.R. Meteorol. Soc. (2002)

8. *p.1618, l.25-26 role of heterogeneous nucleation*

Reply: p.1618, l.25-27 will be changed to: "The observed scavenging ratios may be evidence for the potential importance of heterogeneous ice nucleation for cirrus formation."

9. *p.1619, l.9 role of heterogeneous nucleation*

Reply: p.1619, l.7-12 will be changed to "There are a number of process model sim-

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ulations treating nucleation as a competitive process between homogeneous and heterogeneous freezing (e.g. DeMott et al., 1997). However Sassen and Benson (2000) show that heterogeneous nucleation cannot compete with homogeneous nucleation for most atmospheric situations."

10. p. 1621, l.8-9 temperature range

Reply: OK

11. p. 1633 figure caption

Reply: OK

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

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