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Atmospheric Chemistry and Physics Discussions

ACPD
3, S2379–S2388, 2003

Interactive Comment

Interactive comment on “Thermal stability analysis of particles incorporated in cirrus crystals and of non-activated particles in between the cirrus crystals: Comparing clean and polluted air masses” *by* M. Seifert et al.

M. Seifert et al.

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Reply to reviewer2:

1. Discussion of aerosol number densities

Reviewer: “I would like to see a discussion on the overall numbers of aerosol observed in comparison with other studies.”

Reply: A detailed discussion of the overall numbers of aerosol observed would be beyond the scope of this paper since we are focusing one the thermal properties of crystal residual and interstitial aerosol properties. Note that the relation between residual and interstitial aerosol with respect to number density is subjected in a companion article

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Interactive Discussion
Discussion Paper

(Seifert et al., 2003).

Nevertheless we will change the introduction part of the draft to present results from Minikin et. al. (2003) who investigated the aerosol number densities during the two INCA campaigns.

Action: the second paragraph in the introduction (p.3661 l. 18 to p.3662 l. 6) now reads: “Our knowledge about cirrus from in-situ data is to a large extent based on the FIRE, SUCCESS, EUCREX, ICE, AEROCONTRAIL and INTACC experiments. These projects focused mainly on the Northern Hemisphere (NH) midlatitudes leaving a gap of data in the Southern Hemisphere (SH) midlatitudes. With the project INCA (Interhemispheric differences in cirrus properties from anthropogenic emissions) cirrus observations became for the first time available in the SH hemisphere midlatitudes that allow the comparison of clouds in two with respect to anthropogenic emissions very different regions of the world under comparable meteorological conditions. One of the first results presented from the INCA experiment was a comparison of aerosol number densities in the UT during the two INCA campaigns in cloud free environments (Minikin et al., 2003). As expected, the midlatitudes (50°-60°) of the SH proved to be a rather pristine region, with an average ultrafine aerosol number density of 350 cm-3 compared to 1400 cm-3 in the NH. Refractory aerosol particles, which may act as heterogeneous ice nuclei, were found to be in the order of 35 and 12 cm-3 in the NH and SH, respectively. Based on the observed differences in aerosol properties between the NH and SH in cloud free environments one might speculate if this is also reflected in the properties of the aerosol that is involved in cirrus formation.”

References:

Minikin, A., Petzold, A., Ström, J., Krejci, R., Seifert, M., Velthoven, P., Schlager, M., and Schumann, U.: Aircraft observations of the upper tropospheric fine particle load in the northern and southern hemispheres at midlatitudes, Geophys. Res. Lett., 30, doi: 10.1029/2002GL016458, 2003.

ACPD

3, S2379–S2388, 2003

Interactive Comment

2. Discussion of recent literature:

Reviewer: “The introduction starts with an overview of ice nucleation (lines 10-18 page 3661). However no recent literature is discussed. There are a number of papers by US and European groups discussing the main aspects here and these should be cited in the discussion.”

Reply: ok

Action: We modified the introduction by moving some references used later in the paper to the introduction and added the references suggested by the reviewer. This part of the introduction (lines 10-18 page 3661) now reads:

“This is mainly due to the fact that ice crystals may form through two different processes: homogeneous and heterogeneous nucleation. The relative role of different modes of ice nucleation is still a matter of debate but is thought to critically depend on temperature (DeMott, 2002). Presumably homogeneous nucleation (freezing of a solution droplet) dominates at low temperatures (T< -38°-C), but heterogeneous nucleation (an IN initiates freezing) can become important at higher temperatures, in weaker updrafts or in the presence of large numbers of IN (DeMott et al. 1997; Sassen and Benson, 2000; Kärcher and Lohmann, 2003).”

3. Modification of aerosols when heated

Reviewer: “Pg 3664 line 4-5. The cabin temperature is around 50-70 K warmer than the outside air. This means the aerosol have been heated significantly. Baltenspergers group have shown significant changes in size distribution occur by sampling with a DMA inside a laboratory at a mountain top side as compared to direct measurements at ambient temperature. Others such as Ziemann observe thermal volatilisation of particulate organicat very low temperatures.”

Reply: The reviewer most refers to a study by Nessler et al. (2003) who performed simultaneous dry and ambient measurements of aerosol size distributions at

ACPD

3, S2379–S2388, 2003

Interactive Comment

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Interactive Discussion

Discussion Paper

the Jungfraujoch. The authors found out that the dry total number concentration is often considerably smaller. The particle loss affects almost exclusively the small particles with a diameter D < 100 nm. The average loss of particles with dry diameters D < 100 nm is 35 %. Nessler et al. hypothesis that the loss occurs due to the presence of volatile material, which evaporates during the drying process. Besides ammonium nitrate, volatile organic compounds are expected by the authors to be responsible for the observed particle loss. Nessler et al. hypothesis further that this particle loss is partially due to small, newly formed particles that can only be measured at ambient conditions.

Most in-situ observations of aerosol particles are intrusive in one way or the other, which is especially true for aircraft measurements. Even the ŞambientŤ measurements of aerosol size distribution by Nessler et al. (2003) are intrusive e.g. due to the closed-loop DMA arrangement the ambient RH were slightly delayed. An abrupt RH change occurring at the inlet to the closed-loop system will result in a 50% equilibrium of the RH after 7 min (Nessler et al., 2003).

As an air sample is brought into the aircraft there is always some modification of the aerosol, and therefore aerosol size distributions are normally referred too as ŞdryŤ. With regards to the interpretation of our data it is the particles that evaporated to sizes below the detection limit of the CPC that are of concern. In the temperature range up to cabin temperatures only water vapor can be of significance. If we assume very hygroscopic particles with a growth factor of two, the fraction of particles between 10 and 20 nm in a humid ambient environment may be ŞlostŤ (not detected) when brought into the aircraft. This information is unknown and we can only speculate how large it is based on size distributions and likely growth factors. However, the good agreement between the FSSP-300 and the CVI suggests at least that any underestimation of crystal residual particles can not be serious, unless this underestimation is compensated by some other artifact problem that causes an over-estimation of the residual particles.

The use of the term ŞunheatedŤ (cabin temperature 25-3061616;C) was merely a way to say that the sample lines was not actively heated.

action: We will change the expression “unheated” to “not actively heated”.

4. Limitation of the thermal volatility technique

Reviewer: “Pg 3665 line 1-5: If the particles are com posed of volatile material with a small involatile core, then the CPC/volatility arrangement will detect them as involatile particles, yet the particle properties may be very different from either a particle composed entirely of involatile material. The particle may well have homogeneously nucleated. A discussion of the way the volatility instrument deals with internally mixed particles such as this is vital to the interpretation of the results and may caveat some of your conclusions. You really must introduce these limitations to the technique for the reader to be able to interpret the result for themselves.”

Reply: The mentioned limitations to the volatility technique are discussed, however, first in the discussion section.

Action: We will move the paragraph dealing with this issue to the instrumentation section

5. Data plots: lifecycle of a cloud

Reviewer: “ Page 3666 lines 15-25 and Figure 1 and 2: The authors need to point out how the changes in lifecycle of IN in different clouds affects the superposition of point in the diagrams. In IN rich air many crystals grow quickly suppressing the supersaturation with respect to ice and presumably restricting the points to just over 100% RHi. With fewer ice nuclei for a cloud with the same dynamics and temperature the maximum RHi would increase. A discussion of the variability of this diagram resulting from change in either IN, temperature or dynamics would be useful to guide the reader through. It should be emphasised that RHi is the RH above ice, I believe referee 1 missed this point. It would also be useful to show the variability of Nint on the same style of plot so the reader can assess the ratio of Ncvi to Nint. the plot .”

Reply: First of all we would like to point out that the abbreviation RHi is introduced in the instrumentation section (2.1.3). In literature RHi is commonly referred to as the relative humidity over ice.

In the following we will try to explain the concept of the Ncvi-RHi-diagram in further detail and respond to the reviewers comments dealing with the variablility of IN, dynamics and temperature:

Following an air parcel any cirrus cloud must begin its life cycle in the lower right part of the Ncvi, RHi diagram, because at the point of formation the humidity must be at least above 100% RHi and the crystal nuber density low. At the end of the life cycle the cloud must be in the lower left part of the diagrame, because the relative humidity must be at least below 100% RHi and the crystal number density must be low. Thus we have the starting point and the end point. Exactly, how an individual cloud will move in the Ncvi-RHi diagram from the lower right side to the lower left side depends on variables such as the presence of efficient ice nuclei, water vapor, and uplift velocity, as pointed out by the reviewer.

Although we donŠt know the details there are some general properties about the cloud that might be helpful in interpreting the data. Once ice mass has formed, the cloud starts to deplete available water vapor. This will make the cloud move from right to left in the diagram. A cloud forming few crystals will move close to the base line, whereas a cloud forming many crystals must move up in the diagram and to the left. How far up it reaches depends on mainly the updraft velocity. How strong the component to the left is, depends both of the updraft speed and the ice mass (crystal number density). Thus in the beginning of the lifecycle the cloud increase crystal number density without reducing the relative humidity very much (the relative humidity may even increase if the updraft is strong enough). Once the peak number density has been reached the cloud will move from right to left without changing the number density while the relative humidity relaxes to ice saturation. With this simple cirrus evolution scenario we expect the highest crystal number densities and highest relative humidities to be associated

ACPD

3, S2379–S2388, 2003

Interactive Comment

with the highest updraft velocities (cf Figure 3 the in the companion paper). If all clouds were the same and had the same evolution we would simply have a line moving from lower right, across the figure, and exiting in the lower left. We know that clouds are different, but if there is something like a t't'typical" cloud we would be able to see this in our data. For this purpose the number of observations in each Ncvi, RHi pair was normalized to the maximum number of observations for each given RHi. In other words, along a constant RHi, the maximum number of observations is normalized to one. The results are plotted in Figure 1b in the revised version of the manuscript. If there is a preferred pathway (evolution, life cycle) for cirrus clouds, this should show up in the diagram as a coherent feature where the normalized maxima are linked adjacently. This is also what we find in the frames of Figure 1b as well as in the plots for three other data set (Figures not included).

What regards the reviewers suggestion that we should include a discussion of the variability of the Ncvi,RHi diagram resulting from change in either IN, temperature or dynamics, we would like to point that in this study we are interested of the role that the thermal composition plays in an t't'average" cloud. The fact that we do find a t't'preferred pathway" during cloud evolution shows that there exists something like a t't'typical cloud" despite the large variability in aerosol properties and dynamics. By dividing all the data in two temperature interval we do account for some of the cloud variability, and study the effect that temperature might have on cloud formation. Note that the INCA data set is extensive, but still finite and can therefore not be stratified in as many parameters as one might like to without ending up with insufficient statistics. Furthermore we would like to point out that the relation between Nint and Ncvi is studied in a companion paper (Seifert et al., 2003) where a plot Nint as a function of Ncvi and RHi is provided. Seifert et al. (2003) provides also a plot where vertical wind as function of Ncvi and RHi is expressed.

Action: To make the concept of the Ncvi-RHi diagram more clear we include a new section in the article and remove the paragraph dealing with this concept (p.3666 l.15

to p.3667 l.2)

“3.1 Lifecycle of a cloud

Recently, Ström et al. (2002) suggested a novel way of presenting data that relates an observed parameter to what particular phase of the cloud lifecycle the measurements are performed at. This is simply done by plotting the parameter of interest as a function of relative humidity and crystal number density. Following this concept Seifert et al. (2003) studied observations of vertical wind for the INCA data. Positive vertical velocities were found to be predominately associated with relative humidities above 100 % RHi, whereas negative vertical velocities were found to be predominately associated with relative humidities below 100 % RHi. In this study we plot the frequency of observations as a function of Ncvi and RHi. Figure 1a can be viewed as a probability plot for finding a specific combination of crystal number density and relative humidity. Because there is a very large range in probability the surface is plotted on a logarithmic scale. Nevertheless, a maximum in the observations is located around Ncvi=2 cm-3 and RHi =100%, which is the most probable combination to observe in a cloud. Data at the very lowest relative humidities should be considered with caution since it may arrive from flight segments across cloud edges and where memory effects may cause artifacts.

Following an air parcel any cirrus cloud must begin its life cycle in the lower right part of the Ncvi, RHi diagram, because at the point of formation the humidity must be at least above 100% RHi and the crystal number density low. At the end of the life cycle the cloud must be in the lower left part of the diagram, because the relative humidity must be at least below 100% RHi and the crystal number density must be low. Thus we have the starting point and the end point. Exactly, how an individual cloud will move in the Ncvi, RHi diagram from the lower right side to the lower left side depends on variables such as the presence of suitable ice forming particles, water vapor and updraft velocity.

Although we donŠt know the details there are some general properties about the cloud that might be helpful in interpreting the data. Once ice mass has formed, the cloud

ACPD

3, S2379–S2388, 2003

starts to deplete available water vapor. This will make the cloud move from right to left in the diagram. A cloud forming few crystals will move close to the base line, whereas a cloud forming many crystals must move up in the diagram and to the left. How far up it reaches depends on mainly the updraft velocity. How strong the component to the left is, depends both of the updraft speed and the ice mass (crystal number density). Thus in the beginning of the lifecycle the cloud increase crystal number density without reducing the relative humidity very much (the relative humidity may even increase if the updraft is strong enough). Once the peak number density has been reached the cloud will move from right to left without changing the number density while the relative humidity relaxes to ice saturation. With this simple cirrus evolution scenario we expect the highest crystal number densities and highest relative humidities to be associated with the highest updraft velocities which is consistent with results of Seifert et al. (2003b).

If all clouds were the same and had the same evolution we would simply have a line moving from lower right, across the figure, and exiting in the lower left. We know that clouds are different, but if there is something like a ŞtypicalŤ cloud we would be able to see this in our data. For this purpose the number of observations in each Ncvi, RHi pair was normalized to the maximum number of observations for each given RHi. In other words, along a constant RHi, the maximum number of observations is normalized to one. The result is plotted in Figure 1b. If there is a preferred pathway (evolution, life cycle) for cirrus clouds, this should show up in the diagram as a coherent feature where the normalized maxima are linked adjacently. This is also what we find in the frames of Figure 1b as well as in the corresponding plots for the other data set (Figures for NH warm and SH cold/warm not included).

Having introduced the concept of the Ncvi,RHi diagram we proceed and interpret the thermal composition of the aerosol involved in cirrus formation in the light of the lifecycle of cirrus clouds."

References:

ACPD

3, S2379–S2388, 2003

Interactive Comment

Seifert, M., Ström, J., Krejci, R., Minikin, A., Petzold, A., Gayet, J.-F., Schlager, H., Ziereis, H., Schumann, U., and Ovarlez, J.: Aerosol-cirrus interactions: A number based phenomenon at all?, Atmos. Chem. Phys., 3, 3625-3657, 2003b

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Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3659, 2003.