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Atmospheric Chemistry and Physics Discussions

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.

Received and published: 14 January 2004

2. Modification of aerosols when heated

Reviewer: "The authors describe the thermal stability process: "unheated": cabin temperature 25-30° C; "volatile": 125° C; "semi-volatile": 125-250° C; "non-volatile": >250° C. Cirrus ice, in the cold case, was 235 K (-38° C). By the time the "unheated" measurements are made the residuals and interstitial have been heated at least 63°, 50% of the "semivolatile' temperature range. Urs BaltenspergerŠs group has shown significant modification of aerosols when heated over this temperature range. I would suggest the term "unheated' is not appropriate and some discussion of the changes induced should be made. "

Reply: The reviewer must refer to a study by Nessler et al. (2003) who performed simultaneous dry and ambient measurements of aerosol size distributions at 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. hypothesize 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 “ambient” 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.

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 “dryŠŚ. With regards to the interpretation of our data it is the particles that evaporated to sizes below the detection limit of the CPC that is 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 “lost” (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

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other artifact problem that causes an over-estimation of the residual particles.

The use of the term “unheated” (cabin temperature 25-30°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”.

References:

Nessler, R., Bukowiecki, N., Henning, S., Weingartner, E., Calpini, B., Baltensperger, U.: Simultaneous dry and ambient measurements of aerosol size distributions at the Jungfraujoch, Tellus-B, 55 (3): 808-819, 2003

3. Use of the abbreviations SH/NH

Reviewer: “The terms Northern Hemisphere (NH) and Southern Hemisphere (SH) are used through the Abstract and Introduction. At the beginning of the Experimental section the mission locations, Punta Arenas, Chile and Prestwick, Scotland, are specified. At this point the authors return to the use of NH and SH with blanket descriptions such as pristine, polluted, and clean. I would suggest that NH and SH be replaced by the mission locations. This paper is otherwise misleading in that the reader is drawn to the conclusion that the results are indicative of the entire northern or southern hemisphere whereas measurements were actually only made in two places at one season of the year. Events such as dust storms, biomass burning, and pollution events are hemispheric and seasonal in nature and have an as yet unresolved effect on aerosol number density, lifetime, ice nucleation ability, and cloud formation. I am aware that the use of NH and SH has been used in most INCA publications and the authors are following that lead. The abbreviations are, however, incorrect and prone to misinterpretation and should be removed.”

Reply: We think that the reviewer must agree that the risk that an average reader of ACP will mistake the terms NH and SH for hemispherically averaged properties in an in-situ observation paper must be considered minimal. As for all in-situ observa-

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tion campaigns the measurements provide a snapshot in time and space and strictly speaking only represents the time and space were the observations where made. However, just as the scientific community refers to data as Pacific-, Indian Ocean-, Arctic-, Rainforest-, or tropical-data etc to identify different data sets without necessary confuse these terms with grand averages for these areas, the use of NH and SH should not pose a real problem. There is also some significance to the fact that INCA was the first project to conduct extensive in-situ observations of cirrus clouds in the Southern Hemisphere midlatitudes. Therefore, the two campaigns are not simply the Punta Arenas and Prestwick campaigns or the Clean and Polluted campaigns, it is actually the first time we are able to compare cloud properties in such vastly different ambient conditions. Because the terms NH and SH have been used in a series of papers dealing with INCA data it is becoming more and more intimately connected two the two campaigns. Although, the INCA data it self represents only a month of campaigning at each location, we see no problem in discussing the results in terms of possible hemispheric differences.

Action: The use of NH and SH is short and the comparison approach in so many INCA papers becomes clear, that we see that we can continue using this terminology.

4. Introduction

Reviewer: "The first paragraph of the Introduction describes the formation of ice in the atmosphere, specifically the heterogeneous and homogeneous freezing mechanisms. This topic has been the focus of considerable time by many researchers and yet no references are made here. The authors later reference Karcher, Heymsfield and DeMott. Any of these would be appropriate here, as well as DeMott et al., The susceptibility of ice formation in upper tropospheric clouds to insoluble aerosol components, JGR, 1997. I note that these works underline the non-physical nature of the arguments made in this paper."

Reply: For the comments about the references see our action point below. The state-

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ment that “these works underline the non-physical nature of the arguments made in this paper” we definitely do not agree with. The theories of nucleation are what they are and we do not claim that they are wrong. The question is how applicable they are in the real world, to what extent, where, and when? The reviewer surely agrees with us that the sum of the worldŠs collected in-situ observations that provide information about what types of aerosols that participate in cirrus formation and details around this is meager. Our data suggests that things are perhaps not so simple as one might have hoped the processes in the free troposphere to be. We fail to see why this would disqualify our observations or the theories available. We simply know to little.

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< 235 K), 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, 2002).”

References:

DeMott, P.J., Laboratory studies of cirrus cloud processes, in: Cirrus, Lynch, D.K., Sassen, K., Starr, D.OŠC., Stevens, G. (Eds.), Oxford Univ. Press, New York, 102Ű135, 2002.

DeMott, P.J., Rogers, D.C., and Kreidenweis, S.M.: The susceptibility of ice formation in upper tropospheric clouds to insoluble aerosol components, J. Geophys. Res., 102, 19575-19584, 1997.

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Kärcher, B., and Lohmann, U.: A parameterization of cirrus cloud formation: Heterogeneous freezing, J. Geophys. Res., 108, 10.1029/JD2002003220, 2003.

5. Reference Chen et al. 1998

Reviewer: "The second paragraph of the Introduction describes the previous work by the author showing cirrus crystals controlled by particles smaller than 0.1 mm diameter.Š The authors later reference Chen et al., 1998 which shows a larger heterogeneous mode size with few particles at or smaller than 0.1 mm nucleating ice. These results are inconsistent, yet Chen is used to support the assertion that metallic, crustal, or soot particles are responsible for the observations described in this work. As with the previous point the relation of this work to the relevant literature is lacking."

Reply: The reference to Chen et al. (1998) in this context is perhaps unfortunate but not necessary inconsistent. Chen et al. (1998), clearly state "Thus the particle type abundances that we report should be viewed as qualitative representations of the atmospheric abundances, for that subset of particles larger than about 0.1 μm in size" (p. 1394, second paragraph). The reference to smaller particles is made in brief and the impression is that while analyzing the samples a higher magnification was used once or perhaps a few times. There is no information of what the smallest detectable size is when using the greater magnification, or if more than one sample was analyzed. This is relevant since Chen et al. (1998) states that the composition and abundance of metallic particles varied dramatically from sample to sample.

While the Chen et al. reference can be used to support the presence of IN composed of metalic, crustal, or soot particles it is only representative for a subset of particles larger than ca. 100 nm.

Action: Because of this apparent contradiction the reference Chen et al. (1998) is removed.

References:

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Chen, Y.L., Kreidenweis, S.M., McInnes, L.M., Rogers, D.C., and DeMott, P.J.: Single particle analyses of ice nucleating aerosols in the upper troposphere and lower stratosphere, Geophys. Res. Lett., 25, 1391-1394, 1998

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

Reviewer: "I note that the authors simultaneously published a companion paper in ACP. There is a lead-in to this paper on pg. 3666 (Results, second paragraph) as the authors speculate the connection of their data to the lifecycle of a cirrus cloud. I have three serious reservations äabout this paragraph and the companion paper:

A. The upper third of Figures 1 and 3 are extremely suspect, the remainder at least questionable. This is extended to Figures 3, 5 and 10 of the companion paper. Can the conclusions drawn in the companion paper be justified in this light?

B. The authors discuss the cirrus lifecycle in this paragraph as a combination of RH and Ncvi. No mention is made of the effect of aircraft sampling on this diagram. Specifically, not all clouds have the same maximum crystal density due to formation mechanism (i.e. presence of efficient ice nuclei, water vapor, uplift velocity). Therefore, a cloud with many IN may appear in the lower left, not the lower right, part of the diagram (i.e.,cloud growth occurring in a space defined as evaporation). Likewise, a subvisible cloud might ŠliveŠ only in the lower right portion of the figure (evaporating in the space in which other clouds grow). The observer (i.e., the aircraft instrument) has no means to know the parcel history and, therefore, can not accurately understand what portion of the diagram a mature cloud occupies (see also Figure 3 in the companion paper). How will this effect the conclusions?

C. Data is presented to 20% RH. Can data at less than about 60%, 80% be considered part of a cloud?"

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Reply: A. Without knowing what the reviewer finds “extremely suspect”, we have no direct response to this comment. However, our response to point B might be helpful to the reviewer.

B. The type of figure that the reviewer objects against is a statisical presentation of the observations and not a Lagranian experiment and the reviewer is correct in that we have no means to know the parcel history. However, this is not nessecary and it is possible to interprete the data with respect to a cloud life cycle. Even the reviewer is attempting this in his/her argumentation but makes a few errors.

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

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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 (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 “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 this article. for the two campaigns and the two temperature ranges we used. 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).

C. Although a cloud is something known to everyone, it may sometimes be difficult or even impossible to provide a simple definition for when a cloud is actually present or not. What is the minimum crystal number density or horizontal and vertical extent necessary for an ensemble of hydrometeors to be called a cloud? Is a 1 m thick layer or a particle number density of 1 m-3 sufficient? We can raise similar questions for any observable parameter determined by in-situ or remote sensing methods alike. Because of theses difficulties the presence or non-presence of a cloud is usually determined by the detection limit of the particular sensor used to observe the cloud. What is interpreted as a cloud by one sensor might be interpreted as cloud-free air by another. If we wish to follow the evolution of the hydrometeors it is logical to do so over the humidity range that they exist. Whether this data can be called a cloud or not is completely irrelevant.

Technical Comments:

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1. Reviewer: “ Please correct line 21, page 3668: Š ą Eto an about ą EŠ”
Reply: Do not understand comment. Font style?

2. Reviewer: “Two temperature scales should not be used; please use either K or C.”
Reply: ok
Action: All temperature units will be changed to Kelvin.

3. Reviewer: “ The color scales in Figures 1 and 2 should be consistent; Š hot colorsŠ should represent the same fraction throughout. ”
Reply: We do not agree. Using the same color scales for the volatile, semi-volatile and non-volatile fraction would make it impossible to study trends in the plots. In other words information would get lost for the reader.

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