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Atmospheric
Chemistry
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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.

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1. b) Origin of non-volatile residual particles

Reviewer: “The authors suggest that crustal, metallic, or carbonaceous material may be responsible (pg. 3671). The volume concentration of ice nuclei in layers produced by dust storms may contain up to 3000 per liter (DeMott et al., GRL, 2003) but the number found by the authors can be greater than 2 times this amount. A similar amount of non-volatile material was observed in the interstitial aerosol. To my knowledge no dust events occurred during INCA. Soot has never been found in the upper troposphere within orders of magnitude of these quantities (see the Murphy et al. 1998 reference as well as various studies by J.C. Wilson, P. Sheridan, etc.) especially in the southern

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hemisphere where upper tropospheric air traffic is minimal - where the authors find the largest non-volatile fraction. The authors suggest that sea salt may also be responsible for these particles (pg. 3672). According to various references from INDOEX and ACE there may exist 10 sea salt particles per cc less than 100 nm diameter in the marine boundary layer. If such a parcel is lifted to 100 mb - assuming absolutely no losses such as cloud processing - this leaves Ÿ1 particle per cc. The authors argument would require up to an order of magnitude more, must assume no losses by mechanisms such as precipitation, and is inconsistent with any previous observations in the upper troposphere (see Murphy, Wilson, and/or Sheridan). ”

“I am left to conclude that the most likely reason for the authorsŠ observations is the production of non-volatile material within their own instrument and not an atmospheric phenomenon. The mechanism has been shown to occur by other researchers. The authorsŠ attempts to explain their observations are either incorrect or pure speculation. ”

Reply: The reviewer argues that neither dust, soot, nor sea salt are present in quantities necessary to explain the observations. In the following we will address each of these types of particles.

Dust: The ice nuclei measurements made by (DeMott et al., 2003) were done with the Colorado State University continuous flow diffusion chamber (CFDC). During the dust plume event the CFDC was operating at 123% relative humidity over ice. Since IN concentrations depend on both temperature and humidity (Roger et al., 1998) their results can by no means be viewed as an upper limit. It is plausible to believe that if the CFDC instrument was operating at a higher ice supersaturation more particles would have nucleated. Note that relative humidities over ice above 120% where frequently observed during both INCA campaigns. The dust plume also contains fairly freshly injected dust particles. As the plume ages and dilutes the number density of dust particles naturally decreases, but at the same time water soluble material will have a chance to deposit on the mineral particles. This transformation process most likely also

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improves their ice nucleating ability.

Soot: We would like to point out that the studies referred to by the reviewer are qualitative measurements. Murphy et al. (1998) show examples of ion mass spectra obtained from single aerosol particles in the stratosphere and the upper troposphere, which were typical of the most common spectra. Sheridan et al. (1994) examine the composition and morphology of individual aerosol particles collected during two measurement flights. To our knowledge very few quantitative measurements of black carbon aerosols (BC) in the upper troposphere and lower stratosphere have been reported in literature (Pueschel et al., 1992/1997; Blake and Kato, 1995; Strawa et al. 1999). Note that in the often referenced Blake and Kato (1995) the upper troposphere mid-latitudes are essentially represented by only one data point for each hemisphere (43 °N, 108 °W and 45 °S, 147 °W).

The reviewer is correct in that these observations indeed indicate low mass concentrations. However, more recent measurements (Baumgardner et al., 2003), using a more sophisticated technique than the wire impactors used in previous studies, show quite different amounts of soot particles at high altitude. In the lower stratosphere Baumgardner et al. (2003) show soot number densities between 0.01-30 cm-3 and mass concentrations between 0.3-600 ng m-3 (presentation given on the Aviation, Atmosphere and Climte Conference, Friedrichshafen at Lake Constance 30 June - 3 July 2003, abstract available on http://www.pa.op.dlr.de/pazi/). During the INCA experiments a soot photometer was connected to the CVI. By assuming a specific absorption coefficient (10 m2 kg-1), these measurements provide an equivalent soot mass concentration. From this we can plot the frequency distribution of observed residual mass concentrations for a range of Ncvi between 3 and 10 cm-3. Note that the corresponding plot refers to figure 7 on the following webside: www.itm.su.se/dokument/acpreview.html. Hence, in one cubic meter of ambient air the amounts of soot indicated in this plot is the amount contained in the crystals. Our values are higher than for instance Blake and Kato (1995), but less than presented by Baumgardner et al. (2003). Recall that

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the CVI data is only the absorbing material contained in the crystals. By dividing the soot mass with the number density of residuals we can calculate the mean mass diameter (proportional (σ/Ncvi)1/3 where σ is the amount of absorbing material). Assuming a particle density of 1.5 g cm-3, we get mean mass diameters of 127 nm and 120 nm for the Southern and Northern Hemisphere campaigns, respectively. If the absorbing material is distributed over a distribution of sizes there is room for a fair number of particles below 100 nm.

Sea salt: We are not aware of the publications from the INDOEX and ACE experiment that state what the reviewer says: “According to various references from INDOEX and ACE there may exist 10 sea salt particles per cc less than 100 nm diameter in the marine boundary layer.” However, we know of studies that do indicate large numbers of small sea salt particles. In remote marine air Murphy et al. (1998)* showed that although the sea salt mass fraction of the aerosol was larger at larger sizes, about half the mass near 100 nm was sea salt. These field measurements are in agreement with laboratory simulations by Mårtensson et al. (2003) as well as modeling results by Gong et al. (1997). Since sea salt is produced at the ocean surface by the bursting of air bubbles from entrainment of air induced by wind stress. The number and mass concentration of seal salt aerosol are strongly dependent on wind speed. The mean wind speed in the midlatitudes during fall is clearly different from the mean wind speeds observed during the ACE and INDOEX experiments.

In his/her comments about the composition of the crystal residuals the reviewer makes comparisons as if one of the different particle types must explain all the crystal residues. This is not necessary of course. It is possible that they contribute by some fraction each to the total non-volatile fraction. One possible source of particles that we forgot to mention is extra terrestrial particles such as meteoritic material (Murphy et al., 1998). The volatility observations performed during INCA are the first measurements that at least provide some indirect measure of the chemical nature of Aitkenmode particles present in the tropopause region. There are no other observations that disprove

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their presence.

Actions: P. 3671 line 14-16 now reads: “Aerosol particles acting as heterogeneous IN may be crustal, metallic, carbonaceous material or even meteoritic material (DeMott et al. 2003; Strawa et al. 1999; Sheridan et al., 1994; Murphy et al., 1998), in other words, chemical species that are likely to remain after heating to 250 °C .”

P. 3672 line 25 now reads: “The other possibility is that a significant fraction of the non-volatile particles is made up by NaCl particles. ”

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