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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical analysis of refractory stratospheric aerosol particles collected within the arctic vortex and inside polar stratospheric clouds" *by* Martin Ebert et al.

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To point 1: We have added the study of Renard et al.

To point 2: The deduction of number distributions/mass mixing ratios from single particle data is linked with rather large uncertainties. Nevertheless, we have added the results of our calculation and the discussion of these values (see end of comment). The mass mixing ratio will be around 0.1 ppbm for all nonvolatile particles and the dN/dlogDP for a particle diameter of 0.75 μm is around 10^-2 cm-3. Because of the low number of particles and in this way the high statistical uncertainty, we have resigned to give values for all size bins of all single groups.

New Text in chapter 4:

"In Figure 9 the total number of detected refractory particles for the size ranges 0.5 – 1 μm, 1 – 2 μm, 2 – 3 μm and larger than 3 μm are shown. In principle the number of detected particles can be used to assume an average number and mass concentration of the refractory particles during RECONCILE. As a variety of assumptions has to be made the uncertainty for the received mass and number concentrations is large. For particles smaller than 500 nm no stratospheric concentration ca be received on base of the electron microscopic samples as the large amount of volatile material prevents the determination of the total number of refractory particles on this stage. Considering the total sampled air volume the average mass mixing ratio of the refractory particles (larger than 500 nm) during RECONCILE will be in the range of 0.1 ppbm. The dN/dlogDP value for particle diameters of 0.75 μm is around 10^-2 cm-3. This number/mass loading can be compared with other studies under non-volcanic conditions in the stratosphere. Deshler et al. (2003) published the results of 30 years of in situ stratospheric aerosol size distribution measurements from Wyoming using ballon-borne instruments. He found an average mass mixing ratio for the large mode of 0.02 and in this way 5 times lower as the values determined in this study in late polar winter. Wilson et al. (2008) reports an average mass mixing ratio of 0.4 to 0.8 ppbm for the aerosol sulfate for the non-volcanic influenced time period 1999-2004 and for measurements in the polar vortex only a very low volume of particles larger than 500 nm. Deshler (2008) reports in his review of global stratospheric aerosol measurements about observations of local stratospheric aerosol layers. He mentioned that in the polar regions NAT forms readily on a small fraction (10^-4 – 10^-5) of stratospheric aerosol particles through heterogeneous nucleation. Following this values the fraction of nonvolatile stratospheric particles, which can act as heterogeneous nuclei, is at least 100 times lower as the total number of refractory particles, which we have observed during RECONCILE. He also reports from balloon-borne measurements above the tropopause in Australia, where aerosol layers with particle diameters above 1 μm and number concentrations as high as 10-2 cm-3 were observed. Size and number distribution of this layer seems com-

parable to our findings, whether the possible sources will be completely different (most probably thunderstorms) as in our study. In Weigel et al.(2014) a variety of different model size distributions of stratospheric aerosol are shown. It can be seen that the high number of large particles (>500 nm) is on the upper limit of most model distributions. But for such a comparison it has to be considered, that all presented size distributions do not refer in particular to the situation in the late polar winter vortex. In this way there is the need for further measurements of the stratospheric aerosol characteristics in late polar winter to confirm the observed findings. During RECONCILE Weigel et al. (2014) had observed that the Arctic Vortex in Winter 2009/10 showed an unusually strong subsidence. He found up to 11 submicron particles per cm3 and 8 of these 11 particles were found to contain thermo-stable (250°C) residuals with diameters of 10 nm to 1 μm. Particle mixing ratios (up to 150 of non-volatile particles per milligram air) and fractions of non-volatile particles (up to 75% of totally detected particles) exhibited highest values in air masses having the lowest content of nitrous oxide (70 nmolmol-1 N2O). This indicates that refractory aerosol originates from the upper stratosphere or the mesosphere. He concludes that the impact of refractory aerosol material in the Arctic polar winter is a regular feature. Weigel also estimates the total mass of mesospheric particles deposited over the winter 2009/2010 in the entire Arctic vortex to range between 77 x 10ˆ3 and 375 x 10ˆ6 kg. This estimate yields an enhancement of the refractory aerosol mass by a factor of 4.5 at the lowermost vortex (100 – 67 hPa) toward the end of an Arctic winter. He also compares this value with the expected influx of meteoric material (110 $\pm$ 55 x 10ˆ3 kg/day) and discusses the meaning of non-meteoric sources to the vortex refractory aerosol composition. In the following subchapters we will present the different refractory particle groups, which were detected during the RECONCILE and ESSenCe campaigns by our electron microscopic investigations. For each particle group a discussion about the possible origin and the specific contamination potential is included.“

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