Atmos. Chem. Phys. Discuss., 13, C3911–C3916, 2013
www.atmos-chem-phys-discuss.net/13/C3911/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal cycle and modal structure of particle number size distribution at Dome C, Antarctica" *by* E. Järvinen et al.

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Received and published: 19 June 2013

REPLIES TO REVIEWERS

General

We thank the referee for reading the paper and for the positive comments as well as finding relevant points to be corrected. The relevant changes made in the manuscript are following:

Referee #1

1) We addressed the origin of the nucleation mode particles by adding a chapter 3.4 Origins and atmospheric pathways of particles reaching Dome C during nucleation events. More detailed source analysis will be a subject of a future paper when a longer datasets from the site will be available. As referee #1 pointed out, we also addressed the possible downward mixing of free tropospheric air by calculating trajectories for different heights. When it comes to the issue of ruling out local contamination, we are fairly certain that our treatment of ruling out contaminated wind sectors and too low wind speeds events was adequate to exclude local contamination reaching the measurement instruments. We did some additional study of trajectories to make sure that the air masses have not circulated around the station before event and potentially causing emissions from the station to reach the measurement devices. According to the trajectory analysis in all of the days except on one day, the trajectories are coming far from the station, bringing contamination free air to the station. We also added new chapter to the method section to describe, how we calculated the back-trajectories.

Referee #2

2) As referee #2 pointed out, we added interpretation of our results by adding the new chapter of back-trajectory analysis described above. In additional we added a paragraph in conclusions, where we compared the annual cycle of natural arctic aerosol particles to annual cycle of more polluted arctic particles.

3) We rechecked all the new particle formation events and excluded more events from the analysis. Now we report 60 natural new particle formation events and excluded 21 events. We also remade figure 7.

Detailed answers to reviewers' comments

Referee #1

Page 5735, chapter 2.2.3; figure 9 and table 3: Regarding figure 9, a comparison of the results derived from two different growth rate calculation methods showed tremendous

discrepancies (here: 2.3 nm/h compared to 14.1 nm/h). Please describe in more detail your criteria preferring from case to case a particular method.

In figure 9, the two derived growth rates describe the growth of the mode in two different size classes. In the smaller sizes the growth is more rapid, 14.1 nm/h, but when the nucleated particles reach larger sizes, the growth slows down to 2.3 nm/h. We tried to determine the growth rate of particles under 25 nm with both methods (Hirsikko et al. (2005) and using mean diameter), but the method using mean diameter was not able to fit mode under 20 nm, but the growth rate derived with this method was used to represent the size class between 25-100 nm. In general our methods are limited to derive growth rates from the type of events observed at Antarctica, which means that the errors in our growth rates can be large. The figure capitation in figure 9 is somewhat misleading and was corrected.

Page 5738, lines 22-29 and page 5739, lines 1-14: It should be mentioned that impactor measurements by Udisti et al. were done outside on the roof of the building, while the DMPS was installed inside the (certainly heated) building. Thus I surmise that impactor samples refer to ambient relative humidity (provided the impactor was not heated) in contrast to the DMPS data reflecting the size distribution of (bone-) dry aerosol.

The main point of the lines the reviewer refers to is really just to estimate the order of magnitude of the DMPS-derived volume and mass concentration by comparing it with some relevant numbers published from the site. The difference in the RH on the roof and in the DMPS does not change the conclusion. The higher RH in the impactor on the roof just moves the size distribution somewhat towards larger sizes but does not change the total PM10 mass concentration that was compared here with the DMPS-derived mass concentration. In any case, we'll add some text to the related paragraph – highlighted below – so that it now reads:

... If it is assumed that the particle density is that of water, 1 g cm-3, the mass concen-

trations were 103 ng m-3 and 21 ng m-3 in summer and winter, respectively. With the density of ammonium sulfate, 1.8 g cm-3, the concentrations would be 185 ng m-3 in summer and 38 ng m-3 in winter. It has to be noted here that the samplers of Udisti et al. (2012) were at ambient relative humidity whereas the DMPS sample air got warmer and thus drier when taken into the laboratory air. However, the difference in the RH on the roof and in the DMPS does not change the conclusion. The higher RH in the impactor on the roof just moves the size distribution somewhat towards larger sizes but does significantly not change the total PM10 mass concentration that was compared here with the DMPS-derived mass concentration. The above calculations show that the order of magnitude is the same but detailed comparison needs to be done for a period when both number size distributions and chemical sampling are conducted simultaneously.

Page 5743, lines 13-14 and page 5744, lines 9-18: Calculated growth rates appear astonishingly high, while particle formation rates are particular low. Given the low background particle concentrations, I would expect higher particle formation rates. As stated by the authors, realistic H2SO4 vapour concentrations could explain only a minor part of the growth rate. Do the authors have any ideas about the nature of the condensable gas(es) responsible for the observed particle growth?

More accurate understanding of the growth is not possible, because our measurement methods limit the lower detection limit of the newly formed particles to 10 nm. We could only speculate that the other condensable gases responsible for the growth could be organics, as stated in earlier research papers from Antarctica.

Figure 1: It would be reasonable to use smaller symbols in the third graph (like in the last one) to better distinguish the different traces.

We understand the point raised by the reviewer. The figure was modified according.

Figure 11: I am not really sure whether I understood this plot correctly: I am surprised that your statistics software package generates box-plots in case of a sample size of

just 2! Anyway, to me this does not make much sense. I suggest simply plotting all measured values for each season in the diagrams. This would be a reasonable presentation.

The figure was modified in such fashion that the box-plots are replaced by showing all the values of growth rates and the median growth rate.

Referee #2

Authors use modal fitting procedure. It is not clearly stated in text, but I assume that each size distribution was fitted. With respect to very low aerosol number concentrations, what was the counting statistics error for individual bins and how that could influence fitting results and growth rate calculations? Purely visually I doubt that authors can present growth rates in nm with one decimal precision.

We added error estimates for calculated growth rates. The errors in growth rates were determined as a standard error of the fit to the diameter, time – data. During new particle formation events the concentration of particles is clearly over the noise level, so the counting statistical error has not markedly effect on the growth rate.

Besides DMPS, was there also condensation particle counter measuring aerosol number concentration to which the size distribution integral number can be compared?

No, unfortunately there was only the DMPS for the total number concentration. This undoubtely increases the uncertainty of the results since we cannot calculate the degree of closure between two independent measurement methods. We have now added this to the manuscript.

Chapter 2.2.4 is just repetition from earlier published papers and can be excluded.

We disagree to this and think that it is useful to find the formulas in the method section, rather than have to search them from reference papers.

Page 6, line 21: How many events were actually excluded?

Altogether 21 events out of 81 were excluded.

Page 10, lines 19-21: Neumayer is a coastal station, thus the observed variability might not be due to stronger mixing of the boundary layer (it is not really clear what authors have in mind here), but thanks to stronger influence of the marine air with higher aerosol loading. Check e.g. [Weller et al., 2011]

This is a relevant comment. We have now changed the text on the diurnal cycles so that it reads:

Weller et al. (2011) also detected a diurnal cycle in particle number concentrations at Neumayer. For particles larger than 7 nm this cycle was present for the months September through April but absent from May through August This is not much different from our observations: at Dome C the diurnal cycle of particles larger than 10 nm was strongest in spring (September – November), not quite as strong in summer (December – February) and almost absent in other seasons (Figure 3). Weller et al (2011) noted that the observed diurnal cycle was consistent with a photochemically induced process. The vicinity of the sea with higher aerosol and precursor gas concentrations is probably also affecting the diurnal cycles at Neumayer whereas at Dome C diurnal cycles are most probably only of photochemical origin.

Reference:

Hirsikko, A., Laakso, L, Hõrrak, U., Aalto, P. P., Kerminen, V.-M. & Kulmala, M. 2005: Annual and size dependent variation of growth rates and ion concentrations in boreal forest. Boreal Env. Res. 10: 357–369.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 5729, 2013.