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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Vertical profiling of aerosol particles and trace gases over the central Arctic Ocean during summer" *by* P. Kupiszewski et al.

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We are grateful to the reviewer for his many detailed and useful comments. Our answers to the comments are as follows:

Introduction:

There are a number of existing publications on vertical aerosol measurements over the Arctic using a host of methods. These include both aircraft measurements (e.g. Yamanouchi et al., 2005; Engvall et al., 2008) and unmanned aerial systems measurements (Bates et al., 2013). To the best of our knowledge however, there are

no published results concerning vertical aerosol distributions in the high Arctic using balloons or kites. Furthermore, the aircraft measurements conducted thus far have predominantly taken place at lower latitudes than the ASCOS study, where additional atmospheric mechanisms and processes are of importance. Measurements of vertical aerosol concentration profiles in the high Arctic by helicopter have been conducted as part of the AOE-1996 expedition (Bigg et al., 2001).

The above information will be added to the ACP manuscript.

Instrumentation:

The length of the sampling line is already given in section 2.2.4 (it is 3.05 m). Losses in the line to the UCPC for 10 nm particles are approx. 13%, as calculated using the Particle Loss Calculator (Von der Weiden et al., 2009).

Regarding the temperature difference, as described on p 10405, L25 - p 10406, L2, for the purposes of correction for RH, cabin temperature was assumed to be 20°C, while outside air temperature was measured by the PTU 300 sensor. For the gas phase sampling, the temperature of the sampling line was maintained at approx. 25°C by a self-regulating heating wire.

For the aerosol particles, the temperature change between outdoor and helicopter could cause evaporation and partitioning losses. Currently, the instrumentation does not allow any quantitative measurements of such effects. Considering that the aerosol is transported from mostly below 0°C to approx. 20°C, during a time period of several seconds before the measurement takes place, we estimate that most of the water is removed from the mainly submicrometer-sized aerosol. This should constitute the main mass evaporative loss. Further, any nitrate or ammonia would also (partly) evaporate in this process, however the concentrations in the Arctic are extremely low (Chang et al., 2011). Organics are the main aerosol constituent, which is important for the physico-chemical processing of the aerosol, but the organics are not well characterised. From investigations at lower latitudes, it is known that a change of 20°C induces a substantial

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partitioning loss. We estimate that this does not significantly change the relative size spectra. However, a small bias of sizing towards measured smaller sizes is probable.

With regards to the chemical analysis of DMS and Acetonitrile, the two gas phase compounds we report here, we know that they will almost exclusively be in the gas phase. Their partitioning is negligible even at very high aerosol mass concentrations because of their high volatility.

Estimating C* according to Donahue et al (2006):

C*_DMS (273K) = 5995 ug/m3

C*_ACN (273K) = 584 ug/m3

Example: at 1ug/m3 aerosol mass concentration partitioning coefficients would be 0.017% and 0.17% respectively.

Even at much lower temperatures (and higher mass concentrations) the partitioning would favor the gas phase by far and hence warming the sample up would not release DMS or ACN (because they have not been in the particle phase).

Concerning the aerosol-inlet, it was mounted approx. 1 m in front of the helicopter cabin, so that rotor downwash does not affect the inlet, as long as the helicopter velocity is sufficiently high (sampling true air speed was approx. 19 m/s). We have not checked the effect of the helicopter boundary on the inlet, however we regularly checked that no downwash occurred, based on the onboard aerosol instrumentation.

Results:

In general it is not possible to fly through clouds, due to the presence of super-cooled liquid droplets in the clouds and potential aircraft icing issues. Flight piloting therefore had to be conducted on the basis of visual flight rules (ground visibility had to be maintained), and not instrument flight rules. Typically, the minimum distance between the helicopter and the visible cloud was approx. 100 m. Nonetheless, as a result of

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weather changes and clouds closing up around the helicopter, several flights through the clouds were conducted, such as the flight on the 15th of August 2008, described in section 4.4.

The cases presented are based on common meteorological conditions and air mass origin, as described in section 3.

Measurements of cloud droplet size distributions are not available.

As explained in section 3, the radar reflectivity provides information on the occurrence and size (and hence type) of hydrometeors present. The highest radar reflectivity indicates large hydrometeors (i.e. large ice crystals in snow or cirrus clouds) while lower reflectivity indicates smaller hydrometeors (i.e. cloud droplets, rain or drizzle).

We agree with the reviewer that the manuscript should be considered as focused on aerosols and direct conclusions on cloud properties are not possible. However, we have not made any conclusions on cloud properties; our cloud-related conclusions focus on the origin of CCN relevant for subsequent cloud formation, without directly addressing the properties of the clouds themselves.

Figs. 9a-e: We have not included RH in the individual flight profiles as the general RH profiles are provided for each period on p 10447, Fig. 5b. As the helicopter flights were generally conducted out-of-cloud, often shortly after cloud droplet dissipation, and in conditions where cloud cover is very patchy, the individual flight profiles gives a very localized view of the RH conditions. Therefore, we would prefer to refer the reader to Fig. 5b, and directly describe the RH measured in-flight in the few cases where we refer to it.

The two chemical species are shown for only two cases, as there were no measurements taken for the other flights discussed in detail.

Cloud heights have not been included in the aerosol plots, as the helicopter generally does not fly in cloud. The cloud occurrence and extent, as can be inferred from the

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MMCR radar are presented in Fig. 3b.

Page 10420, line 1ff: The open water is a source for both primary particles (released by bubble bursting), as discussed on p 10419, L19 – p 10420, L 13, but also gaseous species, as mentioned on p 10419, L 13 - 17. Transport of the oxidation products of such precursor gases (e.g. H2SO2 and methane sulphonic acid) from the marginal ice zone (MIZ) can then provide material for further growth of the primary particles emitted in open leads or the MIZ. However, when there is no transport from the MIZ, the concentrations of condensable vapors are very low and particle growth is extremely slow.

Page 1020 line 14 ff: In this case we flew through the cloud. The described cloud processes are not measured per se. However, they are inferred and interpreted based on the cloud properties on the one hand, and the changes in aerosol concentrations throughout the vertical profile on the other.

Page 10420 L 21 – 24: Rephrased to: "As a result, particles within droplets large enough to be lost via settling or impaction in the inlet tubing will not be detected.

Page 10420 L 26 ff: We wrote a general statement here, i.e.: "Generally the Aitken mode particles are not activated in-cloud" – certainly particles as small as 40 nm can be activated in the Arctic, and we note that the concentration of particles in the D14-300 size range, decreases from 200 cm-3 to 130 cm-3. The point we put across here is simply that concentrations of particles in this size range were affected to a lesser degree than those of D>300, as only a portion of Aitken mode particles are activated, as opposed to D>300 particles where almost all particles are activated. In order to make this clearer, we have changed L 26 – 28 to read: "Generally the Aitken mode particles are activated much less efficiently in-cloud, therefore the measured number concentrations of Aitken mode particles should be affected to a much smaller extent by the cloud layer."

In general the flight pattern consisted of perpendicular flights away from Oden, turning

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upwind, and returning. During this time one or several ascents/descents were made. If there were clouds above, we generally flew into cloud base (keeping visual contact with ground below, but with horizontal visibility decreasing significantly). During 12 flights we flew through clouds due to conditions such as weather changes, clouds closing up below us, etc. During these flights we sometimes passed cloud top (August 15 being one such case, as discussed in our article).

We have not noted any increase in 3-14 nm particles (i.e. likely NPF) correlating with above cloud positions for these 12 flights. However, for example the August 31 flight at 23:00 was the first flight measuring NPF, at 400 m altitude , where a cloud had been present earlier until 18:00. Also ground level NPF was measured at 17:20, September 1, approx. 40 min. after fog cleared.

Several flights went further up in clear weather, such that an altitude of 2-3 km was attained several times.

Page 10423 L 5ff: Figure 6c, referred to on p 1023, L 6, shows measurements between ground level and 1200 m. The capping inversion, as stated, is visible in this figure between approx. 600 m and 1200 m, not 400 m and 500 m as the reviewer suggests. We have not marked the height of the low level jet in the figure, as we refer to it only in one case. Therefore we consider it unnecessary to add information on this parameter to the figure, as it would increase the figure's complexity, without providing much relevant information.

Page 10428 L 3 ff: The issue here is whether there is sufficient material for growth of the particles. As discussed in the manuscript on p 10428, L 9 – 17, concentrations of precursor gases were very low (below the detection limit of the PTR-TOF, based on a 30 min integration time). Furthermore, the growth cannot take place via condensation of semi-volatile organic vapors, due to the huge barrier created by the Kelvin effect for 1-2 nm clusters (Karl. et al, 2012).

Page 10428 L 9 ff: This specific case is discussed in detail by Karl et al. (2013); the

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in-text reference has now been added.

We have shortened the period descriptions where possible.

Summary and conclusion:

Page 10433, L 26 – 28: It is not only particles > 300 nm that are absent, but also much smaller particles. While there are particles detected in the 14-300 nm diameter size range, these particles are most likely within the lower range of this size bin. In the TDMPS measurements onboard Oden a mode is visible at approximately 20 – 30 nm, with extremely low particle concentrations at larger sizes. CCN concentration measurements obtained using a CCN counter also confirm the lack of suitable particles for cloud formation, with concentrations of CCN dropping down to 0.3 cm-3 (Mauritsen et al., 2011).

We have included further references to previous studies in order to better demonstrate how the results from this manuscript fit in the scope of previous research.

We have also added recommendations for future measurements required to further our knowledge of aerosol-cloud interactions over the Arctic pack ice.

Technical points: Thanks for noticing the issue with the “double f” – we will make sure this is corrected in the typesetting.

Page 10409, L 5: Liter can be abbreviated with either lower or upper case “L”. The ACPD submission instructions do not specify which is preferable and hence we believe either is fine.

Figure 4: The plots have been enlarged in order to make them easier to see.

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