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Interactive Comment

Interactive comment on "Importance of aerosol composition and mixing state for cloud droplet activation in the high Arctic" *by* C. Leck and E. Svensson

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General Comments:

The authors analyze in great detail a data set of aerosol measurements conducted in the high Arctic onboard the icebreaker Oden during a 3 weeks ice drift in August and September 2008. The measurements are part of the large and interdisciplinary field campaign ASCOS (Arctic Summer Cloud Ocean Study). In this paper the focus is on understanding composition and sources of cloud condensation nuclei (CCN) in the high Arctic. Not much is known about the characteristics of CCN in this remote

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but climatical relevant region. This paper therefore presents potentially valuable and relevant results.

CCN are measured at different supersaturations simultaneously with two DMT CCN counters and aerosol bulk chemical composition (water soluble components) is determined with a 13 stage impactor in 20-40h intervalls. CCN measurments are averaged over the same intervalls and related to the impactor measurements. Impactor measurements are further converted into a chemical number size distribution and combined with measurements of the aerosol number size distribution. The difference between these two (whenever particle numbers exceeded particle numbers determined chemically) is termed "missing non water-soluble fraction". The main focus of the paper is then to infer the properties of the non water-soluble fraction that are needed to achieve CCN closure. This is done by simulating the cloud nucleation process with a Lagrangian air parcel model that includes kinetic effects during water uptake and by modelling the observed CCN activation by Koehler theory. Several assumptions on the missing non water-soluble fraction are made and the ones that seem not to reproduce the observed CCN activation over a whole range of supersaturations are ruled out. Analysis of air mass back-trajectories and estimations of the number of days since the air was in last contact with open ocean are included and a whole picture of CCN composition and sources in the high Arctic is put together.

Structure and Language In general the MS is well structured and figures are ok. However, the main part of the manuscript tends to be overloaded with information. It is hard to follow the core argumentation and the reader gets easily lost. I highly recommend the authors to carefully read over their manuscript and eliminate all information that is not essential. As well, there are several too long and complex sentences that should be rephrased and shortened. I marked some obvious ones in the technical comments below. Check as well for spelling mistakes.

REPLY: The authors thank the reviewer for her/his evaluation of this manuscript, firstly for the "correct" recapture of our intention of this manuscript and secondly for the de-

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tailed and helpful comments. We have addressed all of the reviewer's comments below, each comment in blue text. Changes to the manuscript are marked in "Yellow". We made revisions to improve the clarity of the text, as suggested by both reviewers, by shortening discussions of the results, removing repetitive text and improving the use of English grammar and spelling. A final language check from the ACP editorial office is very much appreciated. We feel that the manuscript have been much improved by the helpful suggestions. The remaining of the above general comments will be covered by our replies to the specific and technical comments made below.

The authors put a lot of effort in the analysis of their results and in the interpretation of the observations. However, the reader gets the impression that the interpretation is somewhat biased. It is a bit disturbing that from the very beginning of the paper the possible presence of polymer gels is mentioned repetitively without ever discussing alternatives. E.g. the extensive interpretation in section 6 seems rather subjective.

REPLY: To date, 23 years after the first Oden expedition (subsequent Oden expeditions in 1996, 2001, and 2008) there are still no other surface aerosol data from the central Arctic pack ice area to compare with. Through these efforts we documented that both areal and seasonal specific meteorological, chemical and biological processes will control the source, formation and fate of the aerosols over the inner Arctic. We have also found out how these processes affected the number size distribution, the chemical composition, and the cloud forming properties of the particles collected.

The fact that near-surface airborne aerosol, as well as low-level cloud and fog droplets, contained the same type of polymer gel material as found in the open-lead surface microlayer strongly suggests an aerosol source within the pack ice. The presence of bubbles in the water column provides a plausible mechanism for getting surface material airborne. This previous work on marine polymer gels and how they resolve over size have most recently been described by Orellana et al. (PNAS, 108, 13612-13617, 2011), Leck et al. (ACP 13, 1-16, 2013), Gao et al. (Ocean Sci. 8, 401-418, 2012) and Karl et al. (GRL, 3738-3743, 2013) and many earlier publications since

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1999.

The unique conditions controling the aerosol life cycle over the pristine and remote inner Arctic is the reason why no attempts have been made to connect our results, with size resolved aerosol number and chemical data and CCN data, to e.g. measurements from the two closest land stations, Mt. Zeppelin, Spitsbergen and Alert, Nunavut. If Leaitch et al., and Engvall et al., would have discussed results on DMS measurements and polymer gels for late summer to autumn conditions the effort could possible have been worthwhile.

Title The paper tries to argue for a possible role of polymer gels in cloud droplet activation in the high Arctic and is not a general discussion of the importance of mixing state for cloud droplet as would be suggested by the title. Change the title for the reason mentioned above.

REPLY: The title is changed to be more specific towards the inner parts of the Arctic. As of to date there are still no other surface aerosol data from the central Arctic pack ice area to compare with, which justify the "review". From past findings we know that atmospheric polymer gels are abundant, but still are occurring both externally and internally mixed with other kind of aerosol types over the inner central Arctic. We therefore do not regard it to be appropriate to be too specific towards polymer gels in the Title. New title: "Importance of aerosol composition and mixing state for cloud droplet activation over the Arctic pack ice during summer".

The possibility of new particle formation by nucleation of precursor gases e.g DMS is not included.

REPLY: We object to that the Leaith and Engval studies have demonstrated new particle formation under similar condition. The DMS source and photochemical sink generating the precursor gases for nucleation (Leck and Persson, 1996a,b; Kerminen and Leck, 2001; Karl et al., 2007; 2011; 2012; 2013) and early growth is both seasonal and temperature dependent. Given that, perhaps the main difference between the studies

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concerns how efficiently nucleation and growth of particles resulting from DMS oxidation are predicted by your choice of model. If the SO2-yield from OH+DMS is high and not constrained by observations sulfuric acid available for nucleation will be too high. The use of constant [OH] during the simulation could give unrealistic high levels of what can be expected in the Arctic. The choice of J and A in nucleation simulations will always be critical to the success of explaining your observations. As shown by Karl et al. (2012) the general rapid appearance of the sub 10 nm particles over the central Arctic Ocean cannot be explained as a result of the growth of the freshly nucleated stable clusters of 1-2 nm diameter by a semi-volatile condensable organic vapor essentially because the Kelvin effect imposes a giant barrier for their initial growth (Karl et al., 2011). In addition, it was shown that the availability of condensable vapors is limited in the boundary layer over the inner Arctic and the concentration of DMS, precursor to H2SO4, is not sufficient to sustain growth into the super 10 nm diameter size range, leaving the observed co-appearance of particles in the 20-50 nm diameter size range coinciding with nucleation unexplained. Thus conventional nucleation paradigms (Karl et al., 2011; 2012) have failed to explain the nucleation phenomenon over the pack ice. An hypothesis explaining the high Arctic nucleation events could be fragmentation and/or dispersion of primary marine polymer gels, 200–500 nm diameter in size, into the nanogel size fractions down to a few nanometer polymers (Karl et al., 2013; Leck and Bigg, 2010); this appears to be consistent with the finding of a particle source in the central Arctic being most pronounced in the smallest particles sizes below 26 nm in diameter (Heintzenberg and Leck, 2012). New text is added for clarification.

Specific Comments:

I agree with Referee #1 that the manuscript needs careful revision before it should be published. The discussion and reasoning are at times unclear and Referee #1 has already raised important points in his comments.

I ask the authors especially to discuss in more detail the uncertainties of their measurements and how they impact the modeling of the CCN activity. The comparison of the

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aerosol number size distribution measured by a TDMPS and the chemical number size distribution obtained by converting mass size distributions measured by the impactor is quite essential for the study. I wonder how significant the differences are between the dsize distributions shown in figure 8:

- Impactor measurement and chemical analysis: Concentrations of species in such a clean environment are very low and challenging to detection limits of instruments.

REPLY: The text in section 2.3 was revised to give a better explanation of the quality of the inorganic analyses of the impactor samples. Based the relative standard deviations derived from the analyses of the blank filters the estimated error in the chemical determinations were between 5 and 20%. The following text was added: Quality checks of the IC-analyses were performed with both internal and external reference samples (organized by EANET, 2008). Systematic errors were less than 2% (with exception for magnesium, Mg^{2+}, with less than 3%) for all ionic components. The analytical detection limits obtained for the various ions, defined as twice the level of peak-to-peak instrument noise, were 0.20, 0.05, 0.01, 0.01, 0.01 and 0.05, 0.00, 0.00, 0.02, and 0.01 μeq dm-3 for ammonium: NH^+_4, sodium: $Na+$, potassium: $K+$, magnesium: Mg^{2+} and calcium: Ca^{2+}, chloride: $Cl-$, MSA: CH_3SOO-, oxalate: $C_2O_4^{2-}$, nitrate: NO_3- and sulfate: SO_4^{2-}, respectively. The overall analytical accuracy was better than 3% and 5% for the anions and cations. Blank levels were determined by loading 6 impactors with: 60 (6 impactors each 10 stages) non-greased substrates and 18 (6 impactors each 3 stages) greased substrates. The blank values for the non greased impactors obtained for the various ions, defined as average and 1σ were 0.20±0.03, 0.13±0.02, 0.02±0.007, 0.02±0.005 and 0.12±0.04, 0.14±0.02, 0.00±0.00, 0.00±0.00, 0.06±0.01, and 0.10±0.01 μeq dm-3 for NH^+_4, $Na+$, $K+$, Mg^{2+} and Ca^{2+}, $Cl-$, MSA, $C_2O_4^{2-}$, NO_3- and SO_4^{2-}, respectively. The corresponding values for the greased substrates were: 0.29±0.03, 0.80±0.16, 0.03±0.004, 0.08±0.005 and 0.29±0.04, 0.22±0.05, 0.00±0.00, 0.00±0.00, 0.10±0.01 and 0.13±0.03 μeq dm-3 for NH^+_4, $Na+$, $K+$, Mg^{2+}

and Ca2+, Cl-, MSA, C2O42-, NO-3 and SO2-4, respectively.

Discuss uncertainties of your chemical measurements and include them in figure 8 and 9. As well discuss the sensitivity of the assumptions you make (e.g. density and hygroscopic growth factor) to convert the mass size distribution into a number size distribution.

REPLY: The uncertainties in the mass to number conversions will be discussed in the text and/or tabulated. To include them in Figure 8 and 9 will not make a readable presentation and therefore they will not be graphically illustrated.

- TDSMPS: the size distribution were averaged over time periods of 20-40h to match the impactor measurements. What was the variability of the size distributions during this time? Include at least standard deviations of the measurements in figure 8.

REPLY: For the reasons given above we will tabulate the statistics on the variability of the TDMPS distributions.

Technical corrections

Below I point out some technical corrections. However, please check your manuscript again carefully for spelling mistakes. REPLY: Done.

P21224 L12-16: Rephrase this long sentence. REPLY: Rephrased.

P21225 L3: should be in particular not particulate REPLY: Changed.

P21225 L4: replace that by than REPLY: Changed.

P21225 L6-10: Rephrase this sentence. REPLY: Rephrased.

P21237 L22: ...mouth of August... change to month REPLY: Changed.

P21238: include the number of DOI in figure 3 REPLY: This information is given in Table 1. We had added this information in the legend of Fig.3.

P21242 L9: eliminate with and : and change to continous; insert with before increasing

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REPLY: Done.

P21242 L21-26: Rephrase long sentence. REPLY: Rephrased.

P21242 L22: change were to where REPLY: Rephrased.

P21243 L18: ...k-values below 0.1 were... not was REPLY: Rephrased.

P 21245 L2: hygroscopic growth factors define of 1.15 ... I don't understand. Should itmean "hygroscopic growth factor of 1.15"? REPLY: Sorry for the mistake, Corrected.

P21246 L17 and 21247 L3: You refer to figure 8c and 8d but the numbering I missing in the figure. REPLY: Sorry for the mistake, Corrected.

P21249 L22: Explain the color coding as well in the caption of the table. REPLY: Done.

P21261 L9: delete of REPLY: Done.

P21260 L25: change humilities to humidities REPLY: Done.

P21261 L2-6: Rephrase sentence. REPLY: Rephrased.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 21223, 2014.