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

Anonymous Referee #2

Received and published: 2 November 2014

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

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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 measurements 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.

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

Title

Change the title for the reason mentioned above.

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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.

Specific comment

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 aerosol number size distribution measured by a TDMP5 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 size 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. 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.

- 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

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this time? Include at least standard deviations of the measurements in figure 8.

Technical corrections

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

P21224 L12-16: Rephrase this long sentence.

P21225 L3: should be in particular not particulate

P21225 L4: replace that by than

P21225 L6-10: Rephrase this sentence.

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

P21238: include the number of DOI in figure 3

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

P21242 L21-26: Rephrase long sentence.

P21242 L22: change were to where

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

P 21245 L2: hygroscopic growth factors define of 1.15 ... I don't understand. Should it mean "hygroscopic growth factor of 1.15"?

P21246 L17 and 21247 L3: You refer to figure 8c and 8d but the numbering I missing in the figure.

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

P21261 L9: delete of

P21260 L25: change humilities to humidities

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P21261 L2-6: Rephrase sentence.

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

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14, C8736–C8740, 2014

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