Reply to 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 #1

General Comments:

This paper uses observations from the 2008 Oden cruise in the high Arctic to examine the cloud condensation nucleus (CCN) properties of aerosol particles sampled near the surface in relatively clean conditions. The CCN observations were averaged over five periods of coincident sampling with size segregated samples of the particles that were integrated over several hours and analyzed by ion chromatography. The results of the IC analyses and further coincident measurements of the particle size distributions are used with an adiabatic parcel model of droplet growth on particles to simulate the CCN number concentrations and compare with the observed concentrations. The authors conclude that the CCN activity is not readily defined by classical Kohler theory. The authors offer an interesting and useful data set. However, as in the comments below, there are a number of issues that must be addressed before the paper is worthy of publication. The paper is very long, much longer than necessary, and while the organization is good, after starting well the writing becomes very difficult in part because of grammatical issues and in part because of repetition. Also, there are several mistakes and spelling errors. Careful editing is needed.

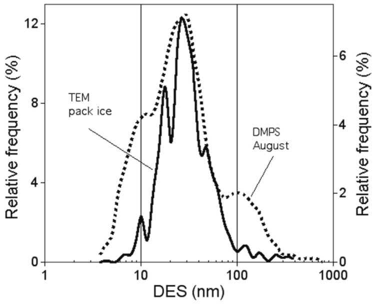
REPLY: The authors thank the reviewer for her/his evaluation of this manuscript and for the detailed and constructive comments. We have addressed all of the reviewer’s comments below, each comment in blue. Changes to the manuscript are marked in “Yellow”. We made revisions to improve the clarity of the text, as suggested by the reviewer, 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 and the presentation of our findings have been greatly improved by the suggestions. The remaining of the above general comments will be covered by our replies to the specific and technical comments made below.

I believe the authors do themselves an injustice by the repeated mention of polymer gels as the source of Aitken particles without objective discussion. That approach makes the paper like an effort at auto-suggestion.

REPLY: To date, 23 years after the first *Oden* expedition (subsequent 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 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 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 1999. Figure 7d (shown below) in Leck and Bigg (Tellus57B, 305-316, 2005) clearly shows a presence of marine gels in the Aitken mode (25-80 nm in diameter).

The unique conditions shaping 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 possibly have been worthwhile, but they did not.



Specific Comments:

1) This is not a “review”. The title needs to be changed.
REPLY: The title is changed to be more specific about 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”. New title: “Importance of aerosol composition and mixing state for cloud droplet activation over the Arctic pack ice during summer”.

2) On page 8, it is mentioned that “During the expedition LPI levels (sample minus blank) of MSA, Cl-, NO3-, SO42, Oxalate, Na+, K+, Mg2+ and Ca2+ down to 0.002, 0.030, 0.009, 0.010, 0.007, 0.030, 0.004, 0.008, and 0.032 nmolm- 3, respectively were detected.” I assume that the units are meant to be “nmol m-3”, which should be corrected. The authors claim exceptionally low detection limits (DL). For example, after blank correction their DL for sulphate is about 0.03 micrograms based on a 30 m3 sample, such as for PI-15. Because the DL are so exceptional as well as critical to the paper, the authors need to discuss their blank sampling further than what is on lines 30-31 of page 7.
How many blanks were collected? What was the average and standard deviation of the blanks for each stage? Was the DL determined using two times or three times the standard deviation of the blanks?
REPLY: To give a better explanation of the quality of the inorganic analyses of the impactor samples the following text was added to section 2.3:
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^{2-}_4, 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^{2-}_4, 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 Ca^{2+}, Cl^-, MSA, $C_2O_4^{2-}$, NO^-_3 and SO^{2-}_4, respectively.

3) Page 4, lines 21-24 - Your discussion seems to separate microgels from other components of the aerosol. When here you refer to organics, are you including marine gels? Please be explicit here.
REPLY: Clarified, see new wording in section 1

4) On lines 7 and 28 of page 4 you refer to the supersaturation range being 0.1-0.7%. On line 9 of page 5, you refer to a range of 0.1-0.9%. In Figures 5 and 6, your range is up to 0.8%. Be consistent.
5) Page 7, lines 8-10 – what do the range of the supersaturations in brackets represent (uncertainty estimates, standard deviations, etc)?
REPLY: After each calibration the settings of the second counter were adjusted. Therefore, the water vapor SS at which the CCNC were measured varies for different time periods. The average values are given in italic and the spread in brackets: *0.10* (0.082-0.106), *0.15* (0.126-0.161), *0.20* (0.171-0.233), *0.41* (0.347-0.521), *0.73* (0.613-0.952) percent with a measurement period of 30 min each. This has now been clarified in section 2.2. Similar to the submitted version of the manuscript we will keep the distinction “average” when

referring to the range 0.1 – 0.7% only in all other cases we will be explicit on the actual range of water vapor SS encountered. The text in section 2.2 (1) was revised accordingly.

6) Page 7, lines 28-29 – Why were only the upper 3 stages greased? Do particles smaller than 3 um not bounce? The specifications for Apiezon-L indicate that it does not dissolve in ketones. What do you mean by dissolved in acetone?
REPLY: The impaction stages 1, 2 and 3 at the inlet of the impactor were coated with grease (Apiezon-L dissolved in hexane) to prevent the bounce-off of larger particles with their relatively large masses onto the downstream stages. The amount of mass, if any, bounced from upper stages is difficult to quantify. However, the fact that sodium concentration in the submicrometer stages did not systematically follow the supermicrometer sodium concentration is an indirect indication that the substrate greasing was sufficient to reduce or eliminate serious bounce-off. The grease was of course not dissolved in Acetone but in Hexane, most sorry for the mistake and thanks. The text in section 2.2 (4) was revised accordingly.

7) Page 11, line 24 – spelling.
REPLY: Corrected.
.

8) Page 12 - Be consistent in labelling of days. In table 1, you use year-month-day. In figure 2, you use day of the year. Use one or the other.
REPLY: The use of "year-month-day" in Table 1 has been replaced with "day of the year ".

9) Page 13 – Lines 1-2 – grammar needs correction.
REPLY: Attempted.

10) Page 13, line 8 – what is "Subjective clustering"?
REPLY: The back-trajectories shown in Fig. 3 b-e, were for the PI-drift subjectively classified in four clusters depending on their geographical origin, that is only taken their origin into account. This has been clarified.

11) Page 13, line 9 and Figure 3 – The figure quality is poor. Zoom in on the plots in figure 3 so that the trajectories are easier to see. None of the trajectories go below 60oN, possibly even below the Arctic Circle.
REPLY: Done.

12) Page 13, lines 19-22 – such a pathway for clean air has also been reported for observations at Alert, Nunavut.
REPLY: Thanks for the information.

13) Page 14, lines 2-4 – ". . .no evidence of any contact. . ."
REPLY: The text is rephrased.

14) Page 14, lines 18-19 – increase in number of CCN or mass in impactor samples? IP or PI?
REPLY: Clarified and corrected in text.

15) Page 14, lines 19-25 – an objective discussion would have acknowledged that other studies have in fact suggested otherwise, e.g. Engvall et al., ACP, 2008; Leaitch et al., Elementa, 2013; Tunved et al., ACP, 2013.
REPLY: These lines are explicitly discussing the properties of measured CCN during ASCOS and not a possible causal relationship between nucleation and CCN.

16) Page 15, lines 10-11 – Why is >300nm a CCN proxy? Smaller particles are more numerous and will contribute more CCN, so why use >300 nm here?
REPLY: The text is rephrased.

17) Page 16, lines 10-14 – There are mistakes in this discussion or in the labelling of Fig 6.
REPLY: Corrected.

18) Page 16, line 16 – should this be OW-1 and PI-8?
REPLY: Corrected.

19) Page 16, line 28 – Page 17, line 2 – this sentence is difficult to understand. Please re-write it clearly.
REPLY: The sentence is rephrased.

20) Section 5.2 and Figure 7 - I cannot see the rationale for the statement on lines 25-27. The use of

"predictions", "expected" and experimentally established" are completely unclear. I assume that the lines in Figure 7 were derived from equilibrium theory, but the apparent data points in Figure 7, which one would assume are the experimentally established values, have the larger diameters. Apparently the experimental results are the lines in Figure 7?
How were the points derived:
a- derive the activation limit dry diameter by comparing the CCN number concentration with that from the TDMPS;
b-estimate kappa from the HTDMA and apply it to the TDMPS measurements;
c- derive critical supersaturations from the measured composition in the impactor samples;
d- other?
REPLY: The points in Figure 7 were derived using the method a) above. The use of "predictions", "expected" and experimentally established" has been clarified both in the text of section 5.2 and in the legend of Figure 7.

21) Figure 7 - What are the units of the diameter axis in figure 7: nanometres? Therange of supersaturation values should be 0.1 to 1, not 1 to 10.
REPLY: Sorry for the mistakes, they are now corrected.

22) Page 18, line 1 - At this point, there is insufficient information provided to make this statement. This goes back to my point in comment 20 above: what information is used to derive the four points (two from PI-10 and two from PI-15) that exhibit activation diameters larger than expected? For point PI-15, figure 2 shows that the CCN concentrations vary between about 10/cc and less than 1/cc. It is quite possible that your chemical composition represents the 10/cc more than your average of 2/cc.

PI-10, figure 2 shows that the CCN measurement covered only a small fraction of the impactor averaging interval. There is no reason to expect that the CCN measurements of sample PI-10 and the chemical composition are sufficiently compatible to declare the CCN to be suppressing the supersaturation.

REPLY: Due to episodes of pollution encountered during sample PI-1 and PI-10 the pumps of the impactors were automatically turned off. During both episodes the temporal records of the TDMPS observations seemed unchanged relative to before their start. We therefore assume no systematic biases in the calculated median CCNC and that the chemical composition of the impactors is sufficiently compatible. This information has been added in the new text. The assumption seems justified by the result by Martin et al. (2011) that also showed that the prediction of the aerosol activation limit dry diameter generally tended to be larger than the expected diameters from the assumed combined pairs of κ-and $\sigma_{s/a}$, when comparing the ASCOS CCN-data at a much higher time resolution than used in this study. Either did the more detailed **inorganic** chemistry allowed for in this study help to explain the “flatness” of the observed SS dependence (Fig. 6). This is why we attempted a best-qualified guess on the missing non-water soluble aerosol fraction based on earlier publications since 1996.

23) Page 18, section 6 - Overall, you converted the EAD to a GMD using an assumed density based on the measured composition, and then you used a HGF of 1.15 from your HTDMA results to reduce the diameters from 50% RH to dry or 20%. What is the variance in the 1.15 factor? Since you later examine scenarios in which you assume different forms of mixing, including external mixing, you need to discuss the impact of the using a constant density of 1.35 and a constant HGF of 1.15 on your later analysis
of the CCN activities.
REPLY: Discussions of these uncertainties are added to text.

24) Page 19, line 9 – “. . .modes POSSIBLY separated. . .” The minimum suggests cloud processing, but that not the only means of developing such a minimum.
REPLY: POSSIBLY added to text.

25) Page 19, lines 14-17 - DMS oxidation products can condense on "smaller, Aitken mode particles". Please include that they can also nucleate new particles.
REPLY: DMS oxidation products are for reasons discussed in the REPLY to comment 29 not likely to nucleate particles over the pack ice area.

26) Page 20, lines 7-9 - The accumulation mode in MIZ-1 is less developed than inPI-10? The peak number concentration in MIZ-1 appears to be close to 200 whereas it just reaches 100 in PI-10. Further the peak inthat mode is at a higher diameter in MIZ-1 than in PI-10. Hence the volume of the accumulation mode in MIZ-1 is larger than that in PI-10. How is PI-10 more developed?
REPLY: Yes we agree. Their accumulation modes are less developed relative to the Aitken mode compared to in sample PI-1. This is now clarified in new text.

27) Page 20, lines 12-15 - Are these sentences supposed to be logically connected? Some re-writing is needed.
REPLY: Rephrased.

28) Page 20, lines 16-17 - why not PI-1?
REPLY: Clarified in text.

29) Page 20, lines 21-29 – Here again objectivity is lacking. It has been demonstrated that under similar conditions that other viable explanations for the new particles exist (see references in comment 15 above).
REPLY: We object to that the Leaitch and Engvall studies have demonstrated new particle formation under similar conditions. The DMS source and the 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 concerns how efficiently nucleation and growth of particles resulting from DMS oxidation are predicted by your choice of model. If the SO_2-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 have failed to explain the nucleation phenomenon over the pack ice (Karl et al., 2011; 2012). 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.

30) Page 21, lines 26-32 – This discussion is very unclear.
REPLY: Rephrased.

31) Page 22, Lines 12-23 – You complicate the calculations here substantially by not using the kappa representation of hygroscopicity. Without kappa, there are too many variables to constrain.
REPLY: We are not sure we understand what is aimed at here. Both the κ-Köhler theory and the kinetic growth equations used in this study require a description of surface tension.

32) Page 25, lines 13-16 – Why couldn't it just be a few particles that had not yet been scavenged, or that had been processed through cloud. Another example of the lack of objectivity in the analysis.
REPLY: Clear sky conditions dominated during PI-15 and the air collected had been advected for 10 days over the pack ice. As discussed in section 4.2 losses of accumulation mode particles to the surface is expected to be at a maximum on the first and second day when drizzle and fallout of fog drops aid deposition to the surface, thereafter losses should have continued by wet deposition at a lower rate, after 10 days of advection most likely showing up as a shoulder of the Aitken mode. This is not what we observed for PI-15. The availability of condensable vapors was also limited with DMS concentrations below the detection limit of 4 ppt(v). Liquid-phase oxidation of SO2 could therefore not have had contributed to the particle accumulation mode particle number population. The limited availability of condensable vapors is also already discussed.

33) Page 25, line 23 – There are no salt particles generated from film drops?
REPLY: 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 (Gao et al., 2012; Leck et al., 2013; Orellana et al., 2011) supports the hypothesis of an aerosol source within the pack ice. The presence of bubbles in the water column (Norris et al., 2011) provides a plausible mechanism for getting surface material airborne. In Bigg and Leck (2008) it was suggested that the highly surface-active polymer gels could attach readily to the surface of rising bubbles and self-collide to form larger aggregates. Consequently, polymer gels and their aggregate production, as well as the embedded solid particles such as bacteria, phytoplankton and its detritus, can be carried selectively to the surface microlayer by rising bubbles. Before bursting, bubbles stay in the microlayer for some time and therefore are likely to acquire walls, consisting to a large extent of strengthening gels, with embedded particulate matter that may be points of weakness as the water drains from between the walls. Following the burst, the film drop fragments would not be drops of salt water but of gel material with salt free water and any particles attached to the fragments. Previous reported result of individual particles by Bigg and Leck (2001, 2008), Leck et al. (2002), and Leck and Bigg (2005a, b, 2010) collected over the pack ice have failed to find evidence of sea salt particles of less than 200 nm in diameter. This supports the suggested mechanism for getting the primary biogenic material at the open-lead surface airborne. This new text is added for clarification.

34) Page 27, line 15 – should "impend" be impede?
REPLY: Yes, indeed. Corrected.

35) Page 28 - How do you initiate growth if you don't allow any condensation when D is equal to D0? In general, there needs to be some discussion of how the modle is initiated.
REPLY: At $D \leq D_0$ the particles are unable to grow in the AC2 simulation case since we assume $\alpha_c=0$. We realize that this is not a realistic assumption; particles are not inert in the atmosphere. However, trying to establish an upper value of α_c that still reduces the particle growth rate sufficiently would be a quite ambiguous parameter-fitting excise. We instead intended to roughly test the idea that the kinetics was restricted rather than to attempt finding the "correct" parameterization. This has now been clarified in the manuscript.

36) Page 28, lines 24-27 – It would save a lot of space if you simply indicated this earlier and went directly to the AC2 scenario.
REPLY: Its not clear what the reviewer means. The discussion in section 9 is directly motivated by the results of section 8. We don't see how one can justify the removal of section 8.

37) Page 28, line 28 to page 29, line 2 - With the formulation for the accommodation coefficient, by the time you get to close to 99% RH your mass accommodation coefficient is effectively unity. What are you evaluating?
REPLY: True for a supersaturated salt solution at 99%RH. But in the case of an aerosol with non water-soluble constituents this is necessarily not the case. Motivations for the assumptions used for the AC-1 and AC-2 simulations are given in section 9. The basic motivation is that we (as well as Martin et al. 2011) failed in reproducing the CCNC at higher water vapor SS even when we tweak the assumption on the chemistry of the "missing non water-soluble fraction". By modifying the condensation accommodation coefficient in section 9 we make yet another attempt to reproduced the CCNC at water vapor SS above 0.2%. The result is encouraging but does not prove cause and effect.

38) Page 29, lines 15-19 – As above, there is so much uncertainty in this analysis, not because it is done poorly, but because there are so many unknowns. Hence, there is no justification for statements such as this.
REPLY: The statement is rephrased to be more balanced.

39) Page 30 – You referred to film drops earlier. What were the wind speeds that led to the bursting bubbles?
REPLY: Norris et al. (2011) have presented the first measurements of bubble size spectra within the surface waters of open leads within sea ice in the central Arctic Ocean. Substantial numbers of bubbles were found

with diameters between 30 and 400 μm. The concentrations are comparable with those observed in the open ocean under modest wind speeds (order of 10 ms^{-1}), but decrease more rapidly with increasing size than for open ocean spectra; no bubbles larger than 560 μm were observed. The total bubble number concentration showed two distinct dependencies on the local environmental conditions: concentrations were highest when the sampled ocean flow has a significant fraction of surface water open to the atmosphere. Concentrations were about an order of magnitude lower when the surface was completely frozen or when the flow was from under the large ice floe, thus isolated from open air for a period of order 10 h or more. With an open water surface, exposed to the atmosphere, the total number concentrations of bubbles also increased with increasing heat flux from the surface to the atmosphere. The observation of significant numbers of bubbles confirms the existence of a plausible mechanism to inject biogenic material from the water surface into the atmospheric surface layer, even in the absence of wind-driven wave breaking. However, still the actual process forming the bubbles is unknown.

Since this is not a conclusion of this work, other potential sources including particle nucleation, need to be considered.

REPLY: 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 primary biogenic aerosol source within the pack ice. This primary source is not only shown to be active in cloud formation over the pack ice but also hypothesized to aid nucleation (Karl et al., 2013). From the data collected during the 4 expeditions in 1991, 1996, 2001 and 2008 to the inner Arctic we have so far never been able to constrain a causal relationship between the nucleation events and chemical properties and state of mixture of CCN with our observations.

40) Conclusions – There is discussion on page 32 that this work indicates a departure from "conventional Köhler theory" for some of the CCN. I disagree. The discrepancies are suggested to be based on kinetics, whereas Köhler equation is equilibrium theory.
The authors have not demonstrated a departure from equilibrium theory.
REPLY: This has been rephrased in the text.

Reply to 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

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 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 detailed 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 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 concerns how efficiently nucleation and growth of particles resulting from DMS oxidation are predicted by your choice of model. If the SO_2-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 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^{2-}_{4}, 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^{2-}_{4}, 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 Ca^{2+}, Cl^{-}, MSA, $C_2O_4^{2-}$, NO^{-}_{3} and SO^{2-}_{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
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 it mean "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.