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Atmospheric Chemistry and Physics Discussions

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 #1

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General –

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.

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.

Comments:

1) This is not a "review". The title needs to be changed.

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?

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.

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)?

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?

7) Page 11, line 24 – spelling.

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.

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

10) Page 13, line 8 – what is “Subjective clustering”?

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.

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

13) Page 14, lines 2-4 – “. . .no evidence of any contact. . .”

14) Page 14, lines 18-19 – increase in number of CCN or mass in impactor samples? IP or PI?

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.

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?

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

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

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

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?

21) Figure 7 - What are the units of the diameter axis in figure 7: nanometres? The range of supersaturation values should be 0.1 to 1, not 1 to 10.

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. For point

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.

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.

24) Page 19, line 9 – ". . .modes POSSIBLY separated. . ." The minimum suggests cloud processing, but that not the only means of developing such a minimum.

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.

26) Page 20, lines 7-9 - The accumulation mode in MIZ-1 is less developed than in PI-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 in that 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?

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

28) Page 20, lines 16-17 - why not PI-1?

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).

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

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.

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.

33) Page 25, line 23 – There are no salt particles generated from film drops?

34) Page 27, line 15 – should "impend" be impede?

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.

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.

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?

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.

39) Page 30 – You referred to film drops earlier. What were the wind speeds that led to the bursting bubbles? Since this is not a conclusion of this work, other potential sources including particle nucleation, need to be considered.

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.

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