Review of "Size-resolved cloud condensation nuclei concentration measurements in the Arctic: two case studies from the summer of 2008" by Zabori et al.

General Comments:

This manuscript presents aerosol number, size distribution, and CCN activity measurements made in Svalbard during June and August 2008. Two, very short (approximately 3-4-day) case studies are discussed, of which portions of these short case studies employ size-resolved CCN measurements for the first time in the Arctic. It is observed that the CCN-derived aerosol hygroscopicity parameter is 0.3-0.4, while bulk chemical composition measurements imply a value of 0.5; the discrepancy is attributed to the dominance of coarse-mode sea salt mass in the bulk chemical composition measurements. NOAA HYSPLIT surface backtrajectories show the airmass history within the marine boundary layer and over the northern, summertime pack ice, but the meaning of these backtrajectories for interpreting the size-resolved CCN data is not clear to me. The same is the case for the micropulse lidar data (Figures 6 and 11), whose main purpose in this manuscript seems to be to show the presence or absence of clouds during the case studies, but which might also be a good tool for characterizing the vertical distribution of aerosols as well. Figures 14 and 15 are the only ones presenting the novel size-resolved CCN measurements, and these are a campaign average for only a single supersaturation (0.4%) and indicate extremely large variability, while the non-size-resolved CCN data in Figures 12 and 13 are discussed and found to be similar/consistent with the literature from the late 1990s and early 2000s. In sum, I am left wondering how these results move Arctic CCN science forward – a more thorough and creative data analysis of the CCN results and/or more extensive size-resolved CCN measurements are required before I could recommend this paper be published in ACP.

Specific Comments:

1) The major limitation of this paper in my mind is that the case study time periods are so short (only 2-3 days each in two summer months of the same year), which makes it hard to draw conclusions regarding the state of Arctic aerosols or CCN in this region from these measurements. Such short case studies might be acceptable if the authors were choosing to analyze / focus on two, unique atmospheric events of unusual significance. For the present manuscript, this does not appear to be the case. Rather, the case study time periods are characterized by modest aerosol and CCN number concentrations (except for a period on June 29), which one might expect for air masses originating in the Arctic, and there is extensive discussion trying to contextualize these very short measurement time periods as fairly typical or even a little higher (~40% during the peak) than those commonly observed (e.g., the discussion regarding the results of Tunved et al., 2013; pg. 5091, Lines 9-30 and elsewhere). Size-resolved CCN measurements have been previously reported for a number of locations worldwide, so the technique is not new. That it is being done for a couple days in the Arctic is the claimed novel contribution of this paper, but I do not think that that in and of itself is worthy of publication. More analysis is required. I suggest the authors explicit address:

    a. What is the new science that is being shown by this study?
    b. How do these results improve (or even affect) our understanding of Arctic aerosol-cloud interactions?

2) Why was only a single supersaturation explored for the size-resolved CCN case? 0.4%SS seems unrealistically high for weakly-forced Arctic clouds. Having data for only one supersaturation makes it much harder to differentiate the aerosol mixing state and size-dependent compositional impacts on the CCN activation spectra. Some past studies have used the combined size distribution and bulk CCN=f(SS) information to estimate the supersaturation-dependent (and therefore size-dependent) kappa values (see, e.g., the already cited papers from the Georgia Tech group). Since there appears to be more non-size-resolved CCN data at multiple supersaturations, it would be perhaps better to focus on these data more extensively (and hopefully for longer time periods as well?).

3) Su et al., ACP, 2010: doi:10.5194/acp-10-7489-2010) proposed a hygroscopicity distribution concept for analyzing size-resolved CCN activity. Padro et al., ACP, 2012 (doi:10.5194/acp-12-10239-2012) also demonstrated methods for the analysis of size-resolved activation spectra. The authors may wish to employ the analysis techniques in these studies in order bolster the analysis section and scientific conclusions in the present work. I would think that running time series of kappa and Dcrit, the non-CCN-active number fraction, and the slope of the activation curve for the time period might allow you to see more clearly if the claimed airmass trajectory changes introduce changes in the aerosol composition and/or mixing state.

4) Figures 14 and 15 are the main size-resolved CCN figures, but they are highly averaged points with a large amount of variability. This variability should be explored more fully as discussed in Specific Points 2-3 above. I suggest that these figures and their associated text be extensively revised and focus more fully on the observed variability and what causes it – e.g., diurnal variability or nucleation events.

5) There are a lot of HYSPLIT and MPL figures that appear to be just cut and pasted into this manuscript, which makes them hard to read and use to interpret the data timeseries. I'd like to see the authors download the HYSPLIT trajectory data and re-plot them in subfigures in Figure 2 and Figure 7. That way it is easier to see what airmass trajectory is occurring at a given time period. Also, it is insufficient to run these once a day – hourly would be better and show the extent to which the air mass origins vary during these time periods. A good example where this has been done well is Figures S1-S3 in the Supplementary Information of Yeung et al., JGR, 2014 (doi:10.1002/2013JD021146).

6) Regarding the MPL figures mentioned in Pt. 4 above, I note that they say "Caution: Preliminary Data", which suggests that these data have not been quality assured for scientific analysis and publication. The authors should confirm that these data are acceptable for this purpose. Also, I'd like to see a more exaggerated scale showing more of the boundary layer features. One thing that is made clear from the backtrajectories is that the surface is decoupled from the 1200 m AGL layer, so the atmospheric state above 10 km has no impact on the surface aerosol conditions explored in this study.

7) On Page 5097, Lines 19-21, it is stated "The presented arithmetic means and SD were calculated assuming a logarithmic distribution of particle concentrations within different particle sizes to

abate the influence of extreme concentrations on the average concentration". I don't know what this means. Arithmetic means and standard deviations are not appropriate for aerosol concentrations and sizes; rather, the authors should compute the geometric mean and geometric standard deviations and replot Figures 2b, 7b, 12, 13, 14 on logarithmic ordinate scales. Also, what are these extreme outliers and where do they come from? If they are due to instrument issues then they should be filtered out during the data QA process, but if they are real then they need to be reflected in the reported results.

8) I don't understand the point of the discussion beginning on Pg. 5098, Lines 16-18: "To the best of our knowledge, to date no size-resolved CCN measurements in the Arctic have been published; therefore, data are compared to results obtained in the subarctic". Is this to say that the Arctic aerosol properties are likely to be similar to those in the subarctic? Also, do the bulk chemical composition measurements suggest the presence of ammonium sulfate, sodium chloride, succinic acid, adipic acid, and glucose in the aerosols (see the discussion on Pg. 5099)? This discussion should be substantially revised to be more relevant to the present study or it should be removed.

9) On Page 5090, Lines 26-28, it is stated that the calculated trajectories do not indicate any precipitation enroute, but this is not apparent from Figures 3-5, 8-10. Please discuss this meteorology information more extensively and how the backtrajectory-met analysis was conducted.

10) On Pg. 5091, Lines 6-8, differences in the air mass trajectories are suggested as a possible reason for the high number densities, but its hard for me to see much of a distinction between the surface trajectory in Figure 4 (when particle concentrations are low) and Figure 5 (when they are high). Perhaps this will be clearer when the revised trajectory figures include more trajectories at hour intervals, but I don't understand this statement as it is presently stated. Is the presence of local pollution another possible explanation?

11) On Pg. 5092, Lines 16-19, it is stated that there are differences in the backtrajectories between Figures 8, 9, and 10. This is true if you follow the air mass back the full 10 days, but the most recent 2-3 days look very consistent between these trajectories and are confined to the surface where production/deposition mechanisms are likely to have a strong influence on the airmass properties rather than long-range transport from far away aerosol sources. Once the figures are revised with hourly trajectories, I'd like to see a more extensive discussion on how meaningful a 10-day trajectory is versus a 3-day trajectory.

12) Pg. 5093, Lines 2-4 states that the Zepplin station is unaffected by clouds, but Figure 11 shows very shallow boundary layers with high backscatter that might indeed suggest the presence of low-level clouds, yes?

13) On Pg. 5101, Lines 18-20, it is stated "In any case, the results suggest that the aerosol resembles ammonium sulphate in its hygroscopic properties and CCN-activation." However, the kappa

values in Tables 1 and 3 indicate that the measured kappa is much lower than that for ammonium sulphate (~0.61). This statement is not right and should be corrected or removed.

14) Figure 12 and 13 do not have to be full page width. Also, they are hard to interpret. What is the meaning of each point? It would be better to see individual spectra connected with lines for the CCN/CN traces. Also, there needs to be some metric for communicating the variability within each of these, presumably averaged, points. It seems to me that percentiles (median and interquartile range) would be best.

Minor Comments:

Pg. 5081, Lines 3-4: What is meant by "active feedback mechanism"?

Pg. 5082, Line 7: I don't think the word "unfavourable" is right here. Sulfate and nitrate salts are highly water soluble and known to act as CCN. Sea salt (NaCl) is indeed more hygroscopic, but it's not clear to me that the difference in kappa between 0.6 and 0.8 is meaningful in the context of Arctic clouds.

Pg. 5082, Lines 8-10: What are the range of kappa values that are being cited here?

Pg. 5082, Lines 16-17: Weird line break

Pg. 5082, Line 21: Do you mean "wet scavenging" instead of "cloud formation"?

Pg. 5082, Lines 27-28 and throughout: Check your significant figures here. Two decimal places on a particle concentration is inappropriate!

Pg. 5085, Lines 23-25: If the results should be interpreted with caution due to the limited number of samples then why are these results being referenced here?

Pg. 5085, Lines 6-7: What is meant by the term "clear proof"? Is this sentence motivating the current study? If so, then the authors should discuss how this paper "provides clear proof".

Pg. 5091, Lines 9-25: Please add these percentile ranges to Figure 2b, so they can be more directly compared with the present measurements.

Pg. 5094, Line 10: These should be geometric means and should have error bars reflecting the geometric standard deviation.

Pg. 5096, Line 28: Clarify where Silvergren et al. made these measurements.

Pg. 5100, Lines 10-13: What is the justification for this assumed surface tension?

Table 2: What are inorganics composed of? Why is a kappa value of 0.53 assumed here?

Figures 2 and 7:

1. The size distribution color scale should be log

2. Please set some non-zero lower limit for the size distribution coloring and make values below this lower limit transparent. This will help the reader see the shape of the distribution given the very low values.
3. It's hard to see the tick marks in part A
4. Make the ordinate axis in part B log.

Figure 6: Is this figure discussed anywhere in the text?

Figure 13: Change CCN/CN scales to be 0 to 1. What is the meaning of that stray point in August?

Figures 14-15: Note that the CCN spectra in these figures are for 0.4% supersaturation.

Figure 15:

1. What does the shaded region denote?
2. Change the ordinate scale to be more reasonable (maybe 0 to 1.1?)
3. Compute geometric mean and standard deviations rather than arithmetic
4. How many activation spectra went into these averaged curves?