Anonymous Review, doi:10.5194/acp-2015-999, Leaitch et al., 2016 ACPD

**General comments:**

The topic of this manuscript is very interesting and worthy of study. The authors provide information on clean liquid clouds in the Arctic that will help us more fully understand the Arctic radiative system, particularly during the under-studied, cleaner summertime environments. The study improves our mechanistic understanding of how aerosols affect these clouds and it provides a needed basis for understanding how changes from anthropogenic aerosols affect the Arctic. This is a topic of relevance and importance. In principle, I think this manuscript may be worthy of publication. However, there are a few changes and clarifications that I would like to see before finally recommending acceptance for publication.

First, the paper could really benefit from a more complete discussion of uncertainties and, in some places, a more thorough statistical analysis. I suggest the authors more fully a) substantiate of some of the claims, b) compare to findings from other works, and c) clarify what is known, not known, and what is just a best guess. The section that was most problematic was the discussion of their methods and assumptions regarding calculating activation size from aerosol concentrations and CCN. I provide more detailed comments and suggestions in the specific comments below.

Secondly, the paper would be more useful to the community if the writing and organization were altered to provide more clarity. I was frequently confused throughout the manuscript about what the authors were doing and why, and I had to re-read some parts multiple times over to understand those parts. Some parts of the manuscript could benefit from more selectively discussing relevant information and eliminating irrelevant information; other parts of the paper would be improved by a more detailed explanation of why the reader should be interested. It might be helpful if the authors could consolidate some of the written information into useful figures or tables. There were also, in my opinion, a high level of references to the supplementary material – to the point that the manuscript does not stand well on it's own. I have given more specific comments and suggestions regarding these points below.

**Specific comments**

*Most important comments:*

1) In the introduction, the authors argue that particles in the 20-100 nm diameter range can become CCN in low aerosol environments. As best I can tell, outside this paper, their initial argument is based only on one previous study (Leaitch et al., 2013). Since a) globally, environments with such low aerosol are fairly rare, b) this hypothesis is not commonly known of or accepted in the community, and c) this hypothesis is a fundamental question/argument to their paper, the authors should describe their reasoning for this argument more fully. For example, if

available, the authors should provide more literature references to support the existence of this process. It would also be helpful to expand on why specifically the findings of the Leaitch et al. (2013) paper support the author's hypothesis. Adding this context will make it easier to evaluate the work.

2) In this paper, the authors make the case that small particles do nucleate cloud droplets based on the following assumptions. First, they assume that the cloud is only incorporating aerosols from below the cloud. Second, they assume that aerosols are preferentially nucleated by size, with larger particles being nucleated first. Thus, they estimate the activation sizes of CCN from the number of CCN in-cloud and the number of aerosols below-cloud in various size classes. However, the various uncertainties in these assumptions need further discussion. What about the impacts of chemistry and hygroscopicity of the individual particles? What about in-cloud aerosol processing or entrainment from above or from the sides? What about precipitation from above? (was there any? Multi-layer clouds are very common in the Arctic). A more clear and straightforward introduction into their assumptions and the uncertainties in these assumptions would be helpful.
3) Other uncertainties that should be discussed more:
    a. Impacts of meteorology on the results. Section 2.3 describes (in way too much detail, I think) the conditions of the study. A lot of information is presented there, but much of it is not directly relevant to evaluation of the key points of the manuscript. At the same time, some very important information is missing, like cloud types (e.g., stratus, cumulus, etc.), cloud sizes, lifetimes, and whether precipitation was occurring. In the summary and conclusions section, it was briefly mentioned that some of the clouds were stratus clouds, but there was no mention of how many. This information should be added sooner and with more detail. There should also be a more thorough discussion of how meteorology and/or surface conditions might or might not affect the results.
    b. Reliability of in-cloud measurements. Were the in-cloud CCN values reliable? What about the aerosol measurements (I know the authors said these were not used in the analysis, but they are shown in Fig. 3)? From what I understand, lot of instruments do not function as well in-cloud as out-of-cloud.
    c. The ability to adequately characterize the influence of smoke from the CO and aerosol data alone (e.g., P.27, l. 595). On P. 14, l. 311 Koellner et al., 2015 is referenced to say that the very slightly elevated CO was from BB. However, I do not think it is appropriate to cite a non-peer reviewed conference abstract to make a point here. Also, CO levels of 100+ ppbv are still quite low compared to other measured CO values in the Arctic, and other summertime studies still count values of CO above this level as "background air" (e.g., Sakamoto et al. (2015, doi:10.5194/acp-15-1633-2015)). Also, particle concentrations were still quite low, weren't they? The authors might consider restating this sentence to mention that the CO levels were elevated compared to your other days, perhaps due to *dilute* BB, but that they were still pretty clean. From what I understand, photochemical destruction of CO would lower average CO values in the summer compared to other seasons, making it

additionally difficult to compare aerosol:CO values to assess influence from biomass burning smoke. Also, there are other source of CO and aerosols besides smoke, and being a gas, CO is an imperfect smoke aerosol tracer to begin with. Anyways, I think the claim that these air masses were influenced by smoke is more uncertain than is currently presented.

   d. P. 24, l. 536: there is an awful lot of scatter (understandably) in figure 11; thus I think there should be some discussion of the uncertainties of the 16 $cm^{-3}$ limit the authors use as their estimate of the Mauritsen limit. Relatedly, on this line I would not call it "the Mauritsen limit", but rather, "our best-guess Mauritsen limit."

4) Other key information that was missing or unclear:
   a. In the introduction/methods there was not a clear, concise description of how the questions would be tested, and how the methods would provide useful information. Please add.
   b. Given that 62 different clouds were examined, it was not clear to me why a select few were chosen for more detailed analysis. Please provide this information.
   c. Why were LA and HA clouds specifically separated at 200 m?
   d. p. 13, l. 292: "for 20 nm particles to activate, the water supersaturations in the bases of the clouds would have had to reach about 1.5%." How was this calculated?

5) There is a heavy reliance on the supplementary figures, and it makes it harder on the reader to have to download another document and scroll through just to understand the main text. It would be better to just put the relevant figures in the main text.

6) I think the authors are actually underselling a couple things:
   a. the fact that no one has observed this Mauritsen hypothesis in the field before. This point makes their work more exciting and I think they could emphasize it more.
   b. the fact that the clouds they are describing are so clean. As noted in the general comments, a better baseline of clean clouds provides a much-needed basis for understanding how changes from anthropogenic aerosols affect the Arctic and this lends importance to the study. This point could also be emphasized more.

*Other specific comments:*

7) Statements that should be backed up more with further statistical analysis:
   a. p. 14, l. 304: "The VMD exceed 20 um compared with a peak value of about 15 um for the July 7 profile." Is this statistically significant difference? Also, please give more information on why this statement is relevant to the paper.
   b. P.22, l. 491: where did you get the 40% value from? Is this statistically relevant?
   c. P.23 l. 503: you say there is a significant difference. What methods did you use to determine the significance, and how significant is it?

8) Other missing or unclear information:

a. P.24, l. 524: "the vertical dashed green line represents the Mauritsen limit below which the cloud may produce a net warming for an increase in the CDNC. " 1) in the earlier text, this limit was estimated as 10 $cm^{-3}$, and in the figure it is estimated at 16 $cm^{-3}$. Since you bring up the figure here first, can you please describe here why there is this difference, and not on line 535, where it is mentioned first? 2) Also, please state why you say in the above text that below this line the cloud can produce net warming. Are you relying on the Mauritsen model results for this statement? If so, please say so.
b. P25, l.558-560: "Variations in other processes, such as mixing or the rate of cooling, may be responsible for the correlation of CDNC and LWC. In these cases, there is sufficient aerosol so as not to be a limiting factor for the CDNC." How do you know that?
c. P.26L l. 577: "The lower- and higher-CO points overlap over a CDNC range of 16 $cm^{-3}$ to 160 $cm^{-3}$." The way this is phrased is a little confusing. I am not sure how it tells you "that 20-100 nm particles from natural sources can have a broad impact on the range of CDNC in clean environments." (l. 579-580)
d. P, 4, l. 81, "remote sensing particle Na do not reflect the size distribution" can you reword to make this sentence clearer, e.g., small particles are not observable, or something like that? I think this could be a very important point for your argument, so it is important to make sure it is clear. (also, please add a reference)
e. P. 11, l. 236: can you explain what you mean by saying that clouds were sampled more frequently during period 1 because of "better visual contrast"? I am confused.
f. Lines 292-300: It is not really explained why the authors are comparing and contrasting the July 7 case with some other unrelated cloud case. Can this be made more clear, or taken out?

9) Abstract: The authors might consider adding a sentence on what this all means for the bigger picture.
10) I agree with Dr. Hudson that the authors should consider using a different to describe cloud droplet number concentration and concentration for cloud condensation nuclei concentration. Currently, the authors use CDNC and CCNC, respectively, and these abbreviations look quite similar, which could lead to some confusion.
11) p. 13, l. 288: "The N30 values compares most closely with the mean CDNC…" [I did not see that in Fig. 3a?]
12) p. 11, l. 246, why are all Na concentrations for particles < 100 nm diameter taken from the SMS, when you also have TSI data down to 5 nm from a different instrument?
13) Did SMS and UHSAS match up fairly well in their overlapping section and generally speaking? Because each might have different artifacts, do you have evidence that these artifacts are either small, or at least similar?
14) p. 12 l. 272: you say it is one of several profiles through the same cloud. How do the other profiles compare?
15) P. 16, l. 351: "The CO mixing ratio is slightly higher in the cloud (81 ppbv) than above (79 ppbv)." I doubt that is statistically significant and outside the error of the instrument. I suggest taking this sentence out.

16) Fig. 8: Are each the points the mean (or median, or something like that) of an individual cloud? Can that be stated more clearly in the caption? Why are clouds within each flight considered separately from each other?
17) p. 21, l. 463: There is no figure 8a. Did you mean Fig. 8? If so, can you be more clear about why LWC and VMD indicate anything for Fig. 8?

*Technical remarks*

1) Fig. 1: can you add an inset that shows Canada, and where Resolute Bay is relative to the larger region? It would also be helpful to show which areas are land vs. ocean.
2) It would be useful to edit Section 3.4.1 for clarity (it is currently 13-sentence paragraph, so breaking it up would help).
3) Section 2.4.1 could be substantially shortened, and made into a table or figure to better elucidate the main points the author wishes to get across. One suggestion would be to add this information on supposed activation sizes into Fig. 3.
4) p. 11, l. 242: "Number concentrations [of particles] greater than 100 nm"?
5) p. 13, l. 289: by, "in the mean " do you mean "on average"?
6) p. 14, l. 302: more variability in what specifically?
7) Fig. 4a: is CO the same axis as altitude, as in Fig. 4b?
8) Fig. S7: Caption says LWC and VMD should be shown, but I only see CDNC
9) Fig. 9: caption says blue, but the points are yellow and black symbols.
10) P22, l48: "The CDNC are plotted against the N50 in Fig. 9b showing that the mean activation size [of HA clouds] was often close to 50 nm."
11) Figure 10: what is this? All the samples? Caption and discussion in the text need to be more clear.
12) Figure 11: you refer to red dots, but I think you meant blue dots
13) Figure S8: do these points represent averages of single clouds? Please specify.
14) P.25, l. 554: "In this low CDNC environment, where cloud droplets may grow large enough to be gravitationally removed from the cloud without the support of collision-coalescence." Reference?
15) P. 25, l.: The reader is referred to the Fig. 11 caption. This information would be more appropriate for a table or for the text.
*16)* P.26, l. 577: The reference of Burkart et al., 2015 is another conference abstract and is not a suitable reference.
17) P. 16, l. 355: "particles [of] about 50 nm"?
18) Table 1 and 2 can be combined into 1 table.
19) P21, l. 475: It is stated that, "As shown in the plot of CDNC versus N100 (Fig. 9a), particles **smaller than** 100 nm activated in most cases", but on p.11 it is said that, "Number concentrations **greater than** 100 nm (N100) are taken from the UHSAS." So on line 475, instead of "smaller than", did you mean "greater than"?
20) P. 27, l. 612: I'd suggest saying ~16 instead of simply 16.