Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-417-RC2, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The role of nanoparticles in Arctic cloud formation" *by* Linn Karlsson et al.

Anonymous Referee #1

Received and published: 4 July 2020

This manuscript presents a set of multi-year measurements of total particle size distributions and cloud residual size distributions at Zeppelin Observatory on Svalbard. This is an impressive and important data set from the Arctic region, with the potential to help better constrain our understanding of aerosol-cloud interactions in Arctic regions. The authors observe sub-100nm (Aitken mode) cloud residuals with some frequency, particularly at cold temperatures in the poorly characterized winter season, and make the claim that these Aitken mode particles play an important role as cloud nuclei in Arctic regions in the coldest seasons.

This is a perplexing and somewhat intriguing result; however, two very major issues arise with this manuscript. First, the impact of measurement artifacts cannot be dismissed in the work. The potential of cloud particle shattering leading to spurious results is discussed in the manuscript (e.g., L251-252, 254-255), and then is almost entirely discounted as a driving factor for the observations of the sub-100nm cloud residuals.

Given the significant uncertainties in these observations, the authors overstate implications of their observations (e.g., L7-9 in the abstract). Second, the manuscript focuses almost exclusively on the small cloud residuals at the expense of other observations, which are also surely valuable and are not given much interpretation. These two issues are elaborated further in the major and specific comments below.

It is clear that while the authors have thought in depth about the possible impact of CVI measurement artifacts, they have not been able to come to any strong conclusions about their impact, and ultimately make the choice to keep all their data in the analysis. My overall suggestion for this manuscript is for the authors to reconsider their focus, and to remove or soften their assertion that sub-100nm particles are important CCN and INP in Arctic winter. This could be accomplished by broadening the scope of the analysis, and particularly the interpretation, to better highlight their observations throughout the year. The authors could take an approach where they first include only data in which they have the highest confidence, and discuss what is learned about aerosol-cloud interactions from those data (i.e., mostly the data collected at warmer temperatures when ice crystal shattering may be less of an issue). The authors could then include the entirety of their data set, in a separate discussion where they lay out the evidence for and against these sub-100nm cloud residuals truly representing the cloud nuclei distribution, making it extremely clear that they cannot rule out measurement artifacts, and providing motivation for future measurements.

Major Comments:

1. The authors provide considerable evidence that their cloud residual Clusters 1 and 2 are associated with ice processes, and use this to suggest that very small particles may be somehow driving ice nucleation. This evidence includes: (1) occurrence at colder temperatures in January and February, (2) high ice occurrence from Cloudnet, (3) association with larger cloud particles, (4) association with times when the cloud residual and cloud particle measurements did not agree well. While all of these are indeed evidence for the presence of ice, they are also evidence for increasing importance

of ice crystal shattering in the CVI, which is a well known issue with this type of cloud residual measurement. Indeed, these measurement artifacts are a partial motivation for developing ice selective inlets (e.g., https://www.atmos-meas-tech.net/8/3087/2015/ and https://www.atmos-meas-tech.net/9/3817/2016/). Further, Cluster 2 is the most frequent, but it shows the most resemblance to a residual distribution you would expect from shattering i.e., nearly uniform across all sizes, bearing little resemblance to the total particle distribution. For this reason, I strongly suggest (as described above) that the authors re-consider the scope of their manuscript to not focus entirely on these smallest cloud residuals.

2. The authors explain their observation of Aitken mode cloud residuals with the possibility of secondary ice formation. However, two issues arise with this interpretation. First, the number of supercooled liquid droplets in a mixed phase cloud should far exceed the number of ice crystals. So, in addition to the small residuals from secondary ice formation there should also be accumulation mode residuals present from supercooled droplets. This does not appear to be the case. Second, INPs are generally not thought to be soluble and are generally larger than a micron (e.g.: https://www.atmos-chem-phys.net/16/1637/2016/ which includes some Arctic data) , and so INP material is unlikely to become fully distributed among secondary ice particles. Another possibility is coagulation scavenging of Aitken mode particles with ice crystals, which could lead to the observed residual size distributions upon shattering and/or evaporation in the CVI. Given these possibilities, the conclusion that Aitken mode particles driven cloud formation in Arctic winter is not supported.

3. An intriguing observation is shown in Figure S7: the comparison between clustered cloud residual size distributions and the total particle size distributions. These demonstrate that when the smallest cloud residuals are present, the total particle size distribution resembles that of the cloud residuals, particularly for for cluster 1. This could be construed as evidence in favour of the author's hypothesis. But, are the total particle sizes measured coming from interstitial particles? i.e., those measured within

a cloud? If so, are these valid particle size distributions, or are they impacted in some way by sampling of cloud particles into the whole air inlet? Do the authors get the same results if they sort the before-cloud and after-cloud size distributions based on the cloud residual clusters? If this result is robust, then I would expect something comparable. Overall, the out of cloud size distributions should be incorporated into this analysis to lend potential support to the conclusions. Further, the data shown in Figure S7 should be shown in the main paper, perhaps combined in some way with Figure 8.

4. Throughout the manuscript the authors appear to confuse the concepts of cloud residuals and cloud nuclei (e.g., L3: "cloud residuals, i.e. particles that were involved in cloud formation", L49-50, the paragraph beginning at L464, L532-535). Cloud residuals are a combination of cloud nuclei, particles that have been effectively scavenged in cloud by droplets and ice crystals, and scavenged particles or cloud nuclei that have been chemically processed within cloud. The one-to-one connection between residuals and nuclei cannot be made. This has direct bearing on the way in which the authors interpret their results.

5. Related to (4) above: the authors also at times appear to misconstrue CCN and INP, which may come from the fact that they cannot distinguish the type of cloud particle they measure. This issue is most prevalent in the interpretation of the results. For example, at L479-480, the authors cite previous studies that have shown small particles can be important CCN in Arctic regions (i.e., when particle numbers are low and supersaturations are high). Given that their observations of small cloud residuals occur in winter when ice processes are important or even potentially dominant, it is unclear how these prior studies directly support their conclusions. Later at lines 486-490 the authors discuss the concept of a CCN-limited cloud-aerosol regime, which is not related to ice formation, and was originally proposed using summertime Arctic observations. While the authors acknowledge this fact in the following sentence, it is not entirely clear how this discussion of the prior literature supports their observations.

6. The authors make frequent assumptions in their analysis that do not appear to have

been arrived at in a quantitative manner. For example on line 230 stating "no correction is preferable to an invalid correction," and the discussion around keeping all data in the analysis at L294-298. Some quantitative assessment of the uncertainty introduced in a correction, versus leaving out the correction, or the impact on the data interpretation when keeping a removing certain suspicious sections of data should be made to build a logical argument for making such decisions. I acknowledge here that making this type of measurement and accounting for all errors is very challenging, and at the same time I hope that the authors will consider this comment thoughtfully when refocusing this manuscript.

7. The monthly average total particle size distributions (orange curves in Figure S8) appear to have a larger dominance of Aitken mode particles in January and February than have been observed in previous multi-year measurements in Zeppelin (e.g., https://www.atmos-chem-phys.net/16/3665/2016/ and https://www.atmos-chem-phys.net/17/8101/2017/). Do the out of cloud, or just before cloud, total particle size distributions look the same as these on a monthly basis? If not, this would suggest that using total particle size distributions measured during cloud events may not be representative of the actual ambient particle populations. How is this observation impacted by the particle loss corrections and density assumptions?

8. Section 2.4: What metrics were used to select the appropriate and physically meaningful number of clusters? Mean euclidean distance? Or any other objective way of looking at optimizing cluster number to explain variability in the data with the smallest number of possible clusters? Four clusters groups the two distributions that contain small particles âĂŤ what evidence is there that these are physically distinct clusters? How was the total particle data incorporated? i.e., the grey size distributions in Figure S7, were they grouped based on the cloud residual clusters? Or clustered separately? If you cluster the out of cloud (I.e., before or after cloud) particle size distributions on their own, do the same five clusters come out?

9. The abstract and conclusions sections contain several statements that are not direct

conclusions from this study. In particular, the conclusions at L 545-547 is not something that can be concluded from observations in this study.

Specific Comments:

L83-93: Observations from satellite are also relevant here, e.g.,: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2012GL053385

L135-140: Is the CVI transmission experimentally determined here explicitly? If so, it should be shown as a function of size in the SI.

L145-179: How well do DMPS 2a and 2b agree in their overlapping size range? How sensitive is the particle loss calculation to the chosen density? A density of 1g/cm3 is likely much lower than the true value.

L165: "Manual screen for outliers and contamination" should be elaborated

L262-267: Would these very low particle concentrations not be consistent with ice clouds in winter? Also, it should not be the absolute amount of particles that matters, but the difference between the two measurements, which is up to 2 orders of magnitude.

L285-293: This appears to be circular logic, and the inability to distinguish between droplets and ice crystals seems to cause a lot of problems here.

L324-327: This could be the case, or could be indicative of a change in supersaturation.

L330-332: Do you expect values up to 2? Does this not give further evidence of shattering, especially at smaller particle sizes? It would be best to include shading around these means to show the range of uncertainty

Figures 3a and 4a: what do the error bars represent?

Figure 4c: why do the authors think that the data for small cloud residuals is not apparent in this analysis?

Figure 6, 7 and 8: These figures are showing a lot of the same information in different ways. This could be focused in such a way to make clear what is most important for the reader to see.

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