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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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Main general comments:

The authors present long term, ground-based measured cloud particle residual number concentration and size distribution results in combination with other aerosol and cloud particle and meteorological data. Like the authors point out, this is indeed a remarkable data set, helping to improve the knowledge about arctic cloud formation.

But:

A. According to the diverse uncertainties (representativity of the measurement location, GCVI and FM-120 sampling efficiency, unassignable assumptions in certain parts of the data interpretation), which are honestly admitted in the manuscript, the conclusions are much too high-flown. Examples are "The reported measurements. . .provide a new basis. . .for developing robust parameterizations of mixed-phase clouds in Earth system models" (line 7-9) or "The direct measurements. . .provide a valuable new perspective

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on Arctic CCN and INP" (line 551-552), although not one INP size distribution or INP concentration is presented in the manuscript. The ball should be kept low with regard to these concluding statements.

B. The main and crucial point of criticism is the discussion and evaluation of the small residual particles (< 30 nm) observed behind the GCVI. The authors try on one hand to find arguments for a cloud physical explanation but on the other hand they admit that artefacts during the measurements cannot ruled out. Unfortunately, they cannot quantitatively estimate the contribution of those artefacts, i.e. is this in the range of 10 or rather 80 %.

C. In the attempt to explain the small residuals scientifically, the authors provide the possibility of small CCN from droplets or "CCN and INP" material from secondary ice particles, without taking a decision for one of the two possibilities or at least provide supporting arguments from other observations. However, for both presented options there are counterarguments, which substantially reduces the chances of their occurrence. Indeed, CCN have been observed as cloud residuals at a diameter of 25 nm in the study of Schwarzenboeck et al. (2000), but the complete cloud residual size distribution and the size dependent activation fraction looked completely different compared to this study, because the larger CCN became activated to a much higher proportion. In general, the reasoning of the authors, that the sampling of secondary ice would result in small cloud residuals is straight forward. But, in a mixed-phase cloud, the supercooled droplets by far dominate the ice particles in terms of number density, i.e. beside the Aitken mode, many more accumulation mode residuals must be present. And why are these secondary ice particles only be observed as small residuals but not as cloud particles by the FM-120 (even if at wrong diameters). Both counterarguments are more indicative for large particle (most likely large ice particles) shattering at or in the GCVI inlet system and subsequent processing of the created fragments. Moreover, in case these small residuals are coming from secondary ice particles, these residuals are in no way involved in the cloud formation process as concluded by the authors.

D. It is known that the results of many airborne CVI measurements in mixed-phase clouds suffer from artefacts by large cloud particle shattering and that beside this effect, the results are hard to interpret with respect to CCN and INP, because such a CVI exactly like the used GCVI cannot differentiate between droplets and ice particles. This was the motivation to develop at least ground-based CVI systems that are able to separate the liquid and iced cloud phases, like the Ice-CVI (Mertes et al., 2007; Kupiszewski et al., JGR, 2016), the Ice Selective Inlet ISI (Kupiszewski et al., ACP, 2015) or the ice selecting pumped CVI IS-PCVI (Hiranuma et al., AMT, 2016). With those systems the residuals of ice particles can be separated and measured in mixed-phase clouds and would be thus needed for the Mt. Zeppelin measurements when mixed-phase clouds prevail.

E. Another drawback is the choice of the cloud particle sensor, which is the major reference instrument for the GCVI in this study. Due to its fixed position its detection efficiency is not clear and moreover it is also not able to distinguish between droplet and ice particles. But for the wintertime measurements and the discussion that the Aitken mode particles are related to ice formation in the cloud a ice particle sensor is crucially needed. It would also shed light into the question whether the measurements are real or mostly caused by artefacts.

Concluding, the manuscript has to be refused in the actual form including the discussion of the wintertime, most likely mixed-phase clouds residual measurements, where the Aitken mode residuals are assumed to be involved in ice particle formation. Even major revisions would not help there.

On the other hand, when the authors could force themselves to restrict their study to the presentation and discussion of pure liquid clouds, their work would indeed provide valuable progress in the understanding of cloud formation in the Arctic.

So, the authors should feel encouraged to change their manuscript in this appropriate way. I do not know if this can be done at this stage of revision or needs a complete new

submission. In hopes that the authors go in this direction, I made a complete review hereafter with minor general comments and specific remarks. Many points are related to the Aitken mode residuals in order to indicate in detail the large doubts of the actual interpretation, but this would then be irrelevant for a revised as requested manuscript.

Minor general comments:

1. Many different number concentrations are elaborated in this study. The authors should think about to use an abbreviation for each of them to make life easier for the reader and themselves.

2. To confirm rather important statements or analysis approaches only references are provided that do not really help the reader. Here the explicit plot in the reference should be given or a plot in the supplement would be highly appreciated. Moreover, some few studies are cited that did not really support the given statement or observation and thus needs intensively revised. These text passages are all mentioned in the “specific remarks”.

3. It is totally unclear why and how the sampling efficiency of an airborne, forward looking CVI are applied to an upward looking ground-based CVI. Since this the basis for the statement that the residual particle concentration agrees rather well with the cloud particle concentration, the applicability of this approach must be justified in the manuscript, which is totally missing.

4. The order of plots in the supplement is not the same as their mention in the manuscript, which should be harmonized.

5. It is unclear whether chapter 3.1 and 3.2 are not so much part of the assessment of the GCVI sampling efficiency but rather much more part of the scientific cloud analysis, maybe except the observation that the GCVI sampling efficiency is reduced at stronger upwind velocities. The authors should think about that.

6. In Addition, it is incomprehensible why the existing ambient aerosol particle data set

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(out of cloud) is not used at all in the analysis. Using short periods before and after cloud appearance, and in comparison with the total aerosol particle data set (inside cloud), the quantitative functionality of the whole air inlet inside cloud could be evaluated. When this is confirmed it would be another approach to derive a GCVI sampling efficiency by scaling the large part in the residual size distribution (> 100 nm) with the total aerosol particle size distribution assuming that those particles are activated with 100%. This result could be again compared with the approaches applied already in this study.

Specific remarks

L.3: This statement is not correct. There are aerosol particles contained in cloud particles due to impaction scavenging or secondary ice formation, which are not involved in cloud formation. So the authors must be more precise and need to reword their description here.

L.4: There is a too drastic change from the research description to the used instrument. This should be done more smoothly, especially not every reader will know a “ground-based counter-flow virtual impactor inlet system” and its peculiarities.

L.6-7: That not only liquid but also mixed-phase clouds exist in the Arctic is known and not only suggested by prior work. Hence, this statement is trivial and should be reworded.

L.16: The reference should be in brackets.

L.20-22: It looks like that something is missing in this sentence. If not, the statement is not totally clear and should be formulated more clearly. What is the cause what is effect?

L.29 and L. 33: What should be the meaning of “typically”? Should be deleted.

L.30: Which scavenging is meant by the authors? Nucleation or impaction scavenging? This is an important aspect and should be mentioned.

L.34: Which particle sinks are meant? The needs to be explicitly listed here for a better understanding.

L.50-51: delete “for example”.

L.55: “performed” should be replaced by “carried out” or something else, since only actors “perform” on stage.

L. 56: not only number concentrations but add also number size distributions.

L.59: it is obvious that the cloud particles are ambient, so this term should be omitted in the manuscript, especially since it could come to confusion with ambient particle size distributions where the term “ambient” is needed.

L.63-64: This listing is redundant to the listing in L.58-59, so the authors should do it completely at one text passage and not two times.

L.116: According to Fig.1 and chapter 2.3.1 only 2 L/min sample flow were indeed used to take the residual particle measurements. Thus, 13 L/min sample flow were not used at all. This is a pity, because when the sample flow would be reduced to the required 2 L/min, the CVI enrichment would be substantially enhanced, which would significantly improve the measurement statistics, which is especially important in the arctic clouds with less cloud particles. The increase in enrichment would increase from about 12 (L.133) to about 90! So, the authors should explain why they did not use this option or if this it is not possible to operate the commercial GCVI with this flexibility.

L.121-122: How save is this trap? Could it be that larger crystals and graupel could break-up in this trap, so that the smaller fragments become resuspended in the system and deliver residual particles in DMPS1? The authors should comment on this and best of all provide a short text passage in the manuscript.

L.124: For the WMO fog definition a solid reference should be given. Moreover, for a reader engaged in cloud physics it would be pretty much appreciated when this value would be transferred and provided as a minimum LWC, which gives a much better

impression about the definition of a cloud in this study.

L. 158-159: Why are two CPC used behind the whole air inlet to measure the total aerosol particle concentration? And the data of which of the two CPC are used in the study?

L. 164: The used particle density is the one of liquid water, but the residual particles are measured at very dry conditions, so the effective particle density is 1.7 g/cm3 or even higher. The same argument holds for the total particles, but due to an expected higher RH in the sampling line, the effective particle density might be 1.5 g/cm-3. The particle density has a rather large influence in particle loss calculations, i.e. these correction calculations has to be repeated and applied to the presented data.

L.165: What is meant by contaminations? The authors need to mention those sources of error explicitly to give the reader a better expression, which situations have been excluded from the study and which not.

L.175: The consequence of the sentence is not quite clear. Does it mean the minor measurement efficiency of DMPS-1 for small diameters was not corrected at all or only not if the concentration in the respective bins was zero. In the presented data, the residual particle size distributions obtained by DMPS-1 often show non-zero concentration down to 10 nm, so that a diffusion loss correction could be in principle applied and it would be also possible to derive a correction from Fig.S2b, which would in both cases better than no correction. Therefore, the authors should say something about this.

L. 183: From Fig.S1 it is obvious that about 2/3 of the cloud sampling was carried out at south wind and 1/3 at north wind conditions. This implies that for 2/3 of the cloud sampling the aspiration efficiency of the FM-120 was very good and for 1/3 of the cloud sampling the aspiration efficiency was pretty bad (since the cloud particles have to make a U-turn to get measured, which will be additionally modulated by the wind speed). Consequently, the first idea would be to do the analysis with the 2/3

south wind cloud cases. The authors should therefore try to go in this direction or present very good arguments, supported by a data evaluation in this respect, why the measurements from all wind directions are equally suited. The mention of only a reference (Koike et al., 2019) is definitely not sufficient.

L. 194: It would be more meaningful to write the METEK user manual explicitly into the brackets and not treat it as a reference citation.

L. 197: Why does the authors did not correct the temperature according to this non-correct temperature difference?

L. 220-222: See comment for L. 183. It should be easy to carry out a FM-120 and GCVI comparison for the 2/3 south wind and the 1/3 north wind conditions to demonstrate whether there is a wind direction or not. This is much better than to "assume" that the fog monitor provides an accurate representation of the cloud particles entering the GCVI. This should be additionally restricted to droplets, since the FM-120 will not correctly measure ice particles.

L. 222-224: The complete GCVI sampling efficiency correction procedure is a very important part of the paper, which the reader must follow more easily. Thus, the mentioned transmission efficiency has to be explicitly shown as a graph in the supplement as only provide the Shingler et al. (2012) reference, especially because the original size dependent efficiency was extended.

L. 227: How dry is the counterflow? This is not mentioned anywhere else.

L. 230: The expression "no correction is preferable to an invalid correction" is pretty unscientific. Should that be a general statement or only related to the concrete approach? If the latter is the case, the authors should somehow quantify why the error is smaller with no correction or should simply remove this expression.

L. 231: The expression "corrected cloud particle concentrations" is badly chosen and misleading. "The GCVI sampled cloud particle concentrations according to the trans-

mission correction" or something similar would make it clearer.

L. 236-238: For the data points below the 1:10 line, it would be very important to know the measured FM-120 cloud particle concentration in order to evaluate how thick the clouds were during these measurements.

L. 247-252: At least the reference of Mertes et al. (2007) is used here in a wrong manner. When there is more than one aerosol particle in or on a ice crystal they will not be emerged as single particles during the drying process in a CVI. These particles will remain on the more and more shrinking ice particle until they will lump together and are released as one particle. Only if the original ice crystal breaks-up the scavenged aerosol particles will be counted individually. This could happen in the cloud (secondary ice) or by hitting any surface of the GCVI. The difference here is that secondary ice would have counted by the FM-120 too, whereas the latter would only be seen as increased concentration in the GCVI, as a shattering artefact, like it is described here.

L. 253: Again a citation, this time Lauber et al. (2018) is used in a wrong sense, because it does not show or even treat the topic of a INP break-up, which indeed is not possible in nature.

L. 263: What is the quantity of "sometimes"? Better change "that sometimes occurs" into "that was observed".

L. 264: The acceleration and deceleration zones are not the only possible locations. Since larger cloud particles could not follow the streamlines, they will simply hit inlet surfaces and walls where the shattering occurs. This needs to be added here.

L. 265-267: Not the concentration but the size of the particles is the important parameter.

L 279-280: The precipitating particles could easily explain the difference in concentrations, since they could also easily shatter at the GCVI surfaces and their fragments are

entering the GCVI and their residuals are counted.

L. 284: Spiegel et al. (2012) only studied liquid droplets and not ice crystals, i.e. this citation is incorrectly used here.

L. 285-293: What is this about now? First the FM-120 was supposed to be correct in order to evaluate the GCVI sampling, now the GCVI concentration results are used to scale FM-120 concentration measurements, and this only when the first is higher than the latter, to account for a FM-120 undercounting. This is dubious and a bit helpless. The much better approach would be to separate the cases with and w/o the presence of cloud ice by an appropriate sensor to prove all these assumptions. Since the amount of ice particles in a mixed-phase cloud is much smaller than the amount of droplets the ice undercounting of the FM-120 would not have much effect.

L. 294-298: This is really a funny conclusion: I cannot really quantify the sources of error in my instruments, therefore I use all the data!

L. 303: The size-dependent transmission efficiency must be shown in the supplement or at least the exact plot in Shingler et al. (2012) must be provided.

L. 309: I would call it “for the sake of convenience”.

L. 324: All particles measured behind the GCVI are residuals. Most likely the authors wanted to say “are in fact CCN”.

L. 324-328: The decisive parameter for this observation at unaltered background aerosol is an increase in cloud supersaturation. This needs to be explicitly mentioned here. A decrease in particle concentration is only a possible cause, just like for example a higher updraft velocity.

L.330-333: a) there should be no uncertainty in the CVI enrichment factor. If so, the authors need to explain this in the manuscript here or before when the working principle of the GCVI is explained. b) The uncertainty of the sampling efficiency would explain a level of the ratio below or above 1 but could not explain the variations of the ratio.

Maybe the counting statistics is not sufficient. The authors, should check for the real reasons for these variations.

L. 339-340: This statement is not true, because when the same size modes would be present in the whole air inlet there would be no peak at 20 nm in Fig.3c. So, Fig.S7 is not a good proof for the given statement, in addition because Fig.S7a also shows this peak.

L.340-341: The statement in L.339-340 (beside the point that it is not really the truth) is not an argument that there are no reasons to expect droplet and even more ice crystal shattering here.

L.343-362: It is totally surprising that there are no residual particle size distributions and ratios residual/total present that show this “ice present effect” of small residuals observed in chapter 3.1. Why is that? These “cold cases” should also be found when the size distributions are sorted by updraft. An explanation by the authors is definitely needed here.

L. 367: Why is no 25th percentile given here, like it is two lines before for the residual number concentration?

L. 375-376: This sentence is not comprehensible. What is meant by “in terms of when peak saturation occurs”? And what is meant by “some differences to previous measurements” in detail?

L.381: The residual particle annual cycle cannot be “confirmed” by the cloud particle measurements. But one could say, that it is “closely related to the cloud particle annual cycle.

L. 393: It is not convincingly proven that this “clear seasonality” is not caused by the artefacts of ice particle shattering. Therefore, I would not use this expression here.

L. 430: What is meant with “total number concentration” in Fig.S11?

L. 436-440: This argumentation supports to some extend the option of large cloud particle shattering as an explanation for the occurrence of the Aitken mode residuals. It is really a pity that no ice particle sensor was operated to bring more light in this problem.

L. 441: Do the authors indeed mean “ambient” particle size distributions (w/o cloud) or is it more likely “total” particle size distributions (within cloud)?

L. 434-448: In contrast to the discussion in the manuscript both Cluster 1 and 2 are most likely an indication for an artefact sampling due to ice particle and large droplet shattering. Cluster 1 are very small Aitken mode particles, but related to a broad size range of cloud particles. The larger cloud particles most likely experience shattering at the GCVI and the very small ones would release only one residual. Maybe this very small fraction might be due to secondary ice formation, but this is not further discussed in detail in this study.

The flat shape of Cluster 2 is a clear indication of large cloud particle shattering, realistically indicating that in this way all particle sizes between 20 and 200 nm occur with more or less the same frequency. At least there is no other mechanism known that could account for such a shape. The existence of artefact measurement by shattering is strongly supported by the fact that this cluster is related to the largest cloud particles measured.

The connection of both clusters with low temperatures and the existence of ice particles further support the occurrence of ice particle shattering artefacts.

L. 464-466: It is very good to point out the difference of CCN and residual particle measurements, but this make only sense when the residuals of liquid clouds are subject of the discussion. Once ice particles occur there is of course a difference since no INPs can be measured in a CCN counter.

L. 467: it is clearer to write “long-term CCN data sets”.

L.479-482: Despite all the caution the authors has exercised in the data analysis by leaving it open whether the occurrence of Aitken mode is an artefact or not, they now claim that the observed Aitken mode particles play an important role for Arctic clouds. But the data situation does not allow for this.

L.486-490: This explanation is incomplete. It explains why smaller particles could become activated, but it does not mention that still the larger ones become activated first. Thus, the accumulation mode particles should be seen much more pronounced, especially because they are activated with 100% and, the 20-30 nm particles only with few % (e.g. Schwarzenboeck et al., 2000).

L. 495-499: Again, the argumentation her is only partly true. Findings from mixed-phase and liquid clouds are mentioned in one sentence, so it is not clear to which cloud type the authors attribute the small Aitken mode particles to. Mertes et al. (2007) indeed measured similar residual size distribution in mixed-phase clouds but with an ice selective CVI, which is not the case in this study. There should be a difference in the measurements since the droplet residuals should be involved here but not are not in the cited study. Consequently, the number of residuals is much less in Mertes et al. (2007), because the CVI was designed to remove large cloud particles that are mostly responsible for artefact measurements. Moreover, the activated fraction showed a strong increase with size, whereas in this study this is not seen (cf. Fig.S7a, Fig.3c). Schwarzenboeck et al. (2000) found droplet activation down to 25 nm particles but at the same time an increase of the activation fraction to 1 above 100 nm, which is totally different to the Aitken mode particle observation in this study and is thus not a confirmation of the findings here.

L. 500-507: It is interesting that the authors do not find an explanation for the occurrence of Aitken mode particles in the Arctic winter. So how should these particles then play an important role in wintertime cloud formation?

L. 508-513: Secondary ice might be possible and would create small residuals, but

the number concentration of ice particles with respect to cloud droplets in mixed-phase clouds is still very low (at least 1 order of magnitude), so that these residuals would not dominate the size distribution. And the Fm-120 would see these particles too, which seemed not to be the case as it was pointed out by the authors. Once more, only CCN but not the insoluble INPs would undergo fragmentation.

L.513-524: Many speculations which are not supported by additional measurements that would have been essential for such a study or using one of the ice selective CVI inlets.

L. 525-528: What are the conditions that are denoted as GCVI malfunctioning and how are they recognized?

L. 529-530: A more scientific procedure would be to remove all data points that could have been biased by potential artefacts and use the "unsuspicious" measurements/results only. Much stronger and first of all justified statements would have been possible.

L.533: Even if one would assume that the small residuals stem from non-shattered cloud particles, the authors always leave it open if these residuals are released from droplets or ice particles. If they are originating from secondary ice (which is to some extend the most likely explanation), these residuals do definitely not contribute to the formation of mixed-phase clouds. The used expression is even in this case a total exaggeration.

L. 538-539: This is a trivial statement that is valid for all clouds around the globe and could be therefore deleted.

L. 540: Before in this study, the separation of Aitken mode and accumulation mode was defined at a diameter of 60 nm, now the authors introduce a size of 100 nm as the lower limit for accumulation mode particles. This is not consistent and has to be harmonized. Since Cluster 1 and 2 are more suspected to present artefact sampling,

the remaining Clusters 3,4,5 are accumulation mode particles according to the first definition, although they are closely to the left edge of the size distribution of the accumulation mode. Hence, it might be advantageous to speak of particles smaller 100 nm instead of declaring that these are Aitken or accumulation mode particles.

L. 543: It puzzles me that the authors claim that the may see the features of a CCN limited regime in their dark period data. Looking at Fig.S7a or Fig.4b it is obvious that the are many particles present in the total aerosol (interstitial + residual) that are not activated. But CCN limited means that there are no more or very few particles to activate, i.e. that there are hardly any interstitial particles left, which is not the case here.

L.544-547: This description is a possible scenario but certainly not a conclusion of this study. Thus, it should be removed or considerably reworded to bring it in the right context to the own conclusions.

Fig.2: Here mean and median are the solid and dotted lines whereas it is vice versa in all other figures. This should be made consistent.

Fig.2: according to the text it is not the measured FM-120 concentration but the GCVI sampled cloud particle concentration derived from the GCVI transmission efficiency, correct? At least that is the explanation in the corresponding text. If so, the figure caption needs to be corrected

Fig. 3 and Fig.4: brackets are use in this way "( ]". In case this has no special meaning, this should be made consistent. Furthermore: The mean, median, percentiles are not mentioned in the figure caption which should be added.

Fig.S4: figure caption: meet instead of meed.

Figures in supplement: Several times the straight lines are named "full", whereas "solid" is the more common term and used in all other figures (supplement and manuscript). This should be made consistent.

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