Reply to reviewers of the manuscript "A long-term study of cloud residuals from low-level Arctic clouds"

Karlsson et al.

March 22, 2021

We thank both reviewers for their helpful and constructive comments. As suggested by the reviewers, we have implemented an individual correction factor of the CVI sampling efficiency based on the concurrent cloud particle measurements. As expected, the overall results were not significantly affected[1] except for the few cases at very cold temperatures when the fog monitor did not sufficiently measure cloud particles. In addition, we added two case studies to the appendix of the manuscript as requested by the reviewer. With these last changes, we hope to have sufficiently fulfilled all requirements and requests. More detailed replies are given below.

1 Anonymous Referee #2

General comments:

With respect to the first version, the actual manuscript has substantially improved. Especially the reduced importance of small residuals with regard to all residual sizes is very good for the work.

However, there remain some critical comments, which are a summary of the main issues of the "specific remarks" below. These specific remarks are more or less only related to content, since language/wording and formal points are very good implemented.

1. Of course, the statistical treatment of the data is unique and very helpful for in-cloud CVI measurements. Nevertheless, two short case studies, one for warm one for cold clouds, where the real physical closure of cloud particle, aerosol particle and residual number concentration are presented would very much increase the confidence in the empirical approach and methodology. Moreover, at least some of the many, many open questions about small residuals in mixed-phase (cluster 1) and warm clouds (cluster 2) could be answered this way or at least some speculations could be ruled out.

The strength of our presented work lies in the availability of a large data set (>1700 hours of in-cloud data), which can be studied using various statistical methods. To address the reviewer's request and thus convey our confidence in our analysis, we have added two case studies in the appendix to the main manuscript. The first case is a straight-forward to analyse liquid cloud case (cluster 5), while the second case is a cluster 1 case that illustrates the difficulties in interpreting data from snowy, ice or mixed-phase clouds. These case studies complement the already

[1] Most figures (very slightly) changed due to the individual-based correction, they are marked with a purple frame in the revised version with track-changes. New figures are marked with a blue box.

existing detailed discussion of our overall observations (with all pros and cons).

As mentioned in the last reply letter, first results from the NASCENT 2019-2020 campaign (see website here) give us now further evidence that clouds at Zeppelin with cluster 1 occurrence (measured by the CVI) are linked to secondary ice processes (Pasquier et al, in prep.). In addition, we would like to mention that the reader can access the data from the Bolin Centre Database and can perform their own individual desired analysis later on.

2. I am still not a fan of the conclusion that say that the measured data could be an artefact or a real physical cloud process or a mixture of both like it still is in the discussion of the small residual particles. In my understanding of research one should avoid or at least quantify artefacts to be sure that they do not exist or at least do not significantly impact my measurements so that the data can then be scientifically interpreted. If this is not the case such data should not be submitted for publication. In this special case it is even more surprising and disappointing, since it is well-known that measurements with "standard", i. e. non-phase segregating, CVI inlets are subject to large artefacts and that therefore several highly sophisticated phase segregating inlets have been developed exactly due to that reason. It is my hope that the authors further reduce their claim that the observed small residuals are related to secondary ice processes in the cloud, because their inlet system is unusable make such a statement.

We need to disagree with the reviewer here. We are grateful for the many constructive suggestions the reviewer has given us (see detailed replies below), but we consider it good scientific practice to discuss potential hypotheses that result from evidence-based findings. We present evidence, both by own analysis and by comparing to literature, that our findings are indeed plausible. At the same time we make the reader aware that certain artefacts can not be excluded and identify/discuss those in great detail. Showing doubts and negative results is an important part of scientific progress. We present and discuss our data and findings in a clear and very balanced way, and no unjustified conclusions are being drawn. Nevertheless, to be more precise, we removed "frequently" from the abstract and are now stating that secondary ice processes is a "potential" explanation for our observations, while clearly stating that artefacts are potentially involved as well. We also added to the abstract and to the introduction the following sentences: "... and the potential contribution of sampling artefacts is discussed in detail." and "Since this is the first long-term deployment of a GCVI inlet, globally and in the Arctic, emphasis will be put on the evaluation of the GCVI inlet sampling efficiency and a detailed discussion of the potential contribution of artefacts during mixed-phase cloud conditions."

We are of course aware that other types of inlets exist that can e.g. be used to uniquely sample ice crystals, which we also address in our manuscript. However, these were not operated or available for our project. Moreover it is unclear if they can be operated on a long-term basis and under the harsh environmental conditions found in the Arctic. In addition, those inlets are unfortunately also not free of artefacts. Last but not least, we would like to state that the here used ground-based CVI is now a commercially available instrument, and as such we find it especially important to present all observations and drawbacks to the reader in a comprehensive and balanced way.

3. I do not know of course what kind of measurement results the authors expected during the long operation period of the GCVI, but it is a pity that the whole set of instruments was not better selected. Beside a phase segregating CVI system, this mainly means a dedicated sensor for droplets (concentration, size distribution, LWC) and ice particles (concentration, size

distribution, IWC). Especially, sensors that are able to measure LWC and IWC would have been much better to clarify the immense amount of speculations and would have been much better as a cloud detector compared to the visibility measurement. In the conclusion, the authors promise to repeat this kind of measurements with an improved set of instruments. This is often done in conclusion chapters without any future action but I hope this time the authors take it serious due to our scientific curiosity about clouds.

Long-term observations in the Arctic at this complexity are a complicated and expensive endeavour which is often dependent on international collaborations. Indeed, our Japanese partners have had installed cloud probes that could have delivered more information about cloud-phase. Unfortunately this particular instrument was not operational during our period. We cannot go back in time and change the instrument set-up, so the best we can do is expand the set of instruments we have and hope that future studies can shed light on the issues where our results were inconclusive. Within the course of this work, we have clearly identified the need for more detailed cloud microphysics observations. Within the recent NASCENT campaign (https://www.aces.su.se/research/projects/the-ny-alesund-aerosol-cloud-experiment-nascent-2019-2020/), several additional instruments have already been installed at Zeppelin Observatory that included a cloud holographic imaging probe (Henneberger et al., 2013). Some of the co-authors are already involved in studies that plan to expand on the analysis presented in this paper. As mentioned above, first preliminary findings seem to indicate the link of cluster 1 to the presence of secondary ice but the detailed analysis is still ongoing.

4. Beside listing all the advantages of the possibility long-term ground-based residual measurements in contrast to short-term measurements with aircrafts, it would be fair to mention the disadvantage in contrast to airborne measurements as well. This means the restriction to clouds with soil contact and orographic effects (which is a general problem and not one at Mt. Zeppelin only) whereas "unbiased" clouds can be reached only by aircrafts. This should be a second point about the representativeness of the measurements together with the one in the site description concerning the representative location of Mt. Zeppelin/Ny Alesund for the Arctic.

We agree and have added to the introduction the following: "Aircraft measurements using CVI inlets have the advantage of recording profiles of undisturbed and elevated clouds but are very expensive and limited in time, while ground-based CVI observations can cover longer time periods (seasons to years) but are potentially affected by the surrounding orography."

5. It is known that the sampling efficiency of CVI systems, and particularly ground-based CVIs, changes with wind and cloud microphysical properties, i.e. it could even change within one cloud event. Therefore, it is nor clear why the authors used only one value to correct for all cloud events included in this study. In principal, each measured residual particle size distribution should be scaled/corrected with the actual CVI sampling efficiency. This is not meant as a harassment, but could indeed lead to different shapes of the averaged residual particle size distributions for the temperature or month intervals (Fig.6b, Fig.7b, and Fig.S8). Consequently, that may also change the complete cluster analysis and the respective discussion. Fig.3a and Fig.5a are not real counter arguments to that because the frequency distribution should be much broader when plotting the ratio value for each residual particle size distribution. Thus, the authors are called to address this issue.

We have reevaluated this part of the analysis and have decided to follow the reviewer's suggestion and implement an individual correction factor for each cloud residual size distribution. We have updated Sect. 3.1.1 and 3.1.2 (now combined into 3.1.1) and other discussion accordingly.

When doing this, we also decided to try to correct for particles lost due to the GCVI cut-size (before we only derived the correction factor above the cut-size) and would like to note here that the average of the new (individual) correction factors is now around 2.2. To our own surprise, this agrees perfectly with the accumulation mode comparison for liquid clouds and gives further confidence in our approach and overall results.

As expected, due to the statistical approach, most figures are dominated by the average correction factor and therefore do not change much when we use individual correction factors (see updated figures). Figures 7 ,8, S8 (now 6, 7, S9), i.e. the figures where data are segregated based on temperature, are the only figures that show a significant change for specific lines at cold temperatures. Since an individual correction depends on how well the fog monitor works, it may introduce an additional bias to the results during conditions when the fog monitor might have issues (e.g. sampling of large cloud particles or ice, or at high wind speeds). We still apply the individual factor, but bring up this potential drawback of that approach in the new appendix (Case II discussion).

Concerning the cluster analysis, we would also like to clear up a misunderstanding by the reviewer. The cluster analysis is not influenced by the choice of correction factor at all, since it is based on normalised size distributions. The correction does of course play a role in the non-normalised size distributions, but because the correction factor (individual or otherwise) is just a scalar applied evenly to each cloud residual size distribution bin, it has no effect on the shape of the cloud residual size distribution.

6. Three times the authors refer to other studies to affirm similar results with their own study (L.569: Verheggen et al. (2007), L. 575-578: Seifert et al. (2003), Mertes et al. (2007)). On the other hand, these studies also show clear differences to the actual study which are not addressed at all (details about that are given in the specific remarks). This is surely not intentional, but needs to be included for a complete scientific discussion.

We have added a couple more sentences that mention how these studies differ from ours (e.g. different background aerosol size distributions and activation ratios). See responses to the specific remarks below.

7. The size distribution of cluster 2 is in this revised manuscript attributed to droplet residuals. But its broad shape is very unusual for a CCN size distribution. This needs to be related to the shape of the total aerosol size distribution simultaneously present at the site. This should be examined during the text passage where cluster 2 is discussed. Moreover, the maximum possible supersaturation should be estimated in order to prove that those small particles can be indeed activated. In principal this should be related to a large updraft velocity, but this seems not to be the case according Fig.10 b.

Cluster 2 is indeed a special cluster that can not be solely described by pure liquid activation. The overall averaged size distributions as such deviate from what would be expected by assuming Koehler theory (see Fig. 12 and the discussion within the manuscript). As described in more detail below, there are several studies now that give clear evidence that 25-80 nm particles can be activated for liquid cloud cases (see e.g., Bulatovic et al., 2021) and as such we see no need to perform own calculations. However, we have added a few more sentences to the revised manuscript that describe the total particle size distributions of cluster 2: "The cluster is bimodal, but relatively broad and flat. While the same approximate size modes are present

in the total particle size distribution, the average cluster 2 cloud residual size distribution has a less pronounced minimum and slightly lower concentration of accumulation mode particles than the total particle distribution. The shape of cluster 2 could perhaps also be influenced by ice processes (i.e. not all cloud residuals correspond to CCN), or the shape might be affected by evaporation of volatile compounds from the accumulation mode particles. However, this cannot be confirmed without size-resolved chemical composition or volatility measurements, which were not available for our period."

I would have preferred to completely forego the presentation and discussion of the residual particle size distribution measured during mixed-phase conditions (mainly the cluster 1 discussion with 8 % of the time) and I still have some doubts about the actual existence and interpretation of cluster 2. However, this is certainly not enough to reject this manuscript presenting a unique long-term cloud particle residual data set at an important place on earth with respect to the role of clouds to arctic amplification. The authors seem to stick to these points, which is acceptable in the way it is implemented, although it does not strengthen the manuscript to my opinion. However, the issues raised in the general comments and the specific remarks in this review must be responded and where appropriate included in the manuscript for publication.

Specific remarks

L.59: beside secondary ice one should at least also mention impaction scavenging as an in-cloud process that could result in residuals that are not identical to the original CCN or INP.

We agree, we have added this to the sentence so it reads "[...](e.g. impaction scavenging or secondary ice [...])".

L.60-64: Already here it would be fair to mention the disadvantage of ground-based or advantage of aircraft in-situ cloud measurements as already mentioned in my general comments.

We added the following sentence here: "Aircraft measurements using CVI inlets have the advantage of recording profiles of undisturbed and elevated clouds but are very expensive and limited in time, while ground-based CVI observations can cover longer time periods (seasons to years) but are potentially affected by the surrounding orography."

Figure 2b,c; Fig.3a,b; Fig.4a; Fig5a,b: Maybe it is given overseen by me, but it is important to indicate the averaging time of the data points.

In almost all cases, data were averaged to the resolution of our main instrument (i.e. the DMPS behind the GCVI). We apologise that this was not stated more clearly. We have added the following in the introduction of the Results section "Unless otherwise stated, all data presented in this section have been averaged to match the time resolution of the cloud residual size distributions measured by DMPS 1 (i.e. 5–7 min averaging time, cf. Tab. 1). When making simple comparisons to DMPS 2, which has a lower resolution than DMPS 1, we used all simultaneously measured data (i.e. overlapping DMPS scans, without repetition of data points). In the cases where a one-to-one data point comparison was necessary, both DMPS data sets were downsampled (usually to 30 min averages).".

As for Fig. 4, it uses 30 min averages. This is stated in the text but had been forgotten in the caption, so we added "30 min mean values of DMPS 1 and DMPS 2b data were used for this analysis" to the caption.

L.383-389: Here the argumentation of the authors is not correct. At this text passage clouds are discussed with a cloud particle concentration of 1 cm-3 and where the cloud particle concentration is below the cloud residual concentration. These must be more or less totally glaciated clouds, where most cloud particles (ice crystals) are rather large. This means, the very likely shattering of these ice particles in the GCVI wind tunnel would substantially increase the cloud residual but not the cloud particle concentration. This is supported by many airborne CVI measurements in ice clouds.

This is a good point. As discussed in the text, cloud particles would need to exceed $\sim 70\,\mu$m to be likely to shatter in the wind tunnel. We believe the reasoning still stands that for mixed-phase clouds, where such large crystals would be far outnumbered by droplets, potential shattering would not significantly influence the cloud residual concentrations. We added "mixed-phase" into the sentence to clarify ("[...]in most mixed-phase non-precipitating clouds, the concentration of large cloud particles is much lower than the total cloud particle concentration[...]").

If the clouds are fully glaciated, we agree with the reviewer that artefacts may come from cloud ice as well as precipitating ice. We changed the end of the paragraph to: "Therefore, the magnitude of the concentration difference we observe suggests that precipitating particles (e.g. snow) are a more likely cause than large cloud droplets or ice crystals. If, on the other hand, we are sampling a fully glaciated cloud consisting of large ice crystals, then shattering artefacts could come from cloud ice as well as precipitating ice."
Having said that, the only instrument we have to try to verify the cloud particle size and concentration is the FM-120, which has an upper size limit of $50\,\mu$m. In other words, if the pure ice clouds should consist of very large crystals, those crystals might not be detected by the fog monitor. The visibility comparison, while not perfectly valid for ice clouds, did show that the fog monitor may have issues detecting the particles. However, cloud particles that are so large would perhaps also be too large to enter the GCVI (without artefacts). All in all, with our instrumental set-up we cannot clearly disentangle these effects.

L.413-416: This analysis is very crude. The match of the total aerosol and cloud residual size distribution, which is more or less the exact and individual knowledge of the GCVI sampling efficiency for each cloud included in the respective data set, has to be taken into account and not only a correction factor of 2 for all clouds. This is important, because the value of the D50% is very sensitive to the correct quantitative relation of total particle to residual particle size distribution. For a first guess that can be done by normalizing the plateau between 100 and 300 nm to 1. Doing so, one would see a trend in Fig.6c, i.e. a decrease of D50% for higher updraft velocities. This would be the expected behaviour, because this means higher supersaturation with the capability to activate smaller particles. Thus, this approach leads to contrary conclusion with regard to the actual text and should be at least commented by the authors, since this is a crucial point for the description of the properties of warm arctic clouds.

Because we are deriving the $D_{50\%}$ values from average size distributions, the behaviour is dominated by the average correction factor. Using an individual correction factor does not significantly change the relative behaviour of the curves in this figure. Nevertheless, we have decided to follow the reviewer's suggestion and implement an individual correction factor (see response to comment 5 above and comment L.425-426 below for more details).

We agree with the rest of the comment, and have made a figure where the ratios have been normalised to 1 at 200 nm, see Fig. 1a. Figure 1b shows the interpolated $D_{50\%}$ values from the normalised ratio for each updraft interval (together with the size range of the diameter bin in

which the value falls). This figure is included in the supplementary material and discussed in the main text of the revised manuscript as follows: "The $D_{50\%}$, defined as the diameter where the ratio is 0.5, ranges between approximately 57 and 75 nm in Fig. 5c. One can attempt to account for the aforementioned sampling issues by normalising the plateau of the ratios to 1. If this is done, the $D_{50\%}$ ranges from 50 to 78 nm and shows a decreasing trend with increasing updraft (Fig. S8). This behaviour is expected from a cloud physics point of view, where higher updraft velocities can produce higher supersaturation levels which, in turn, allows smaller particles to activate."

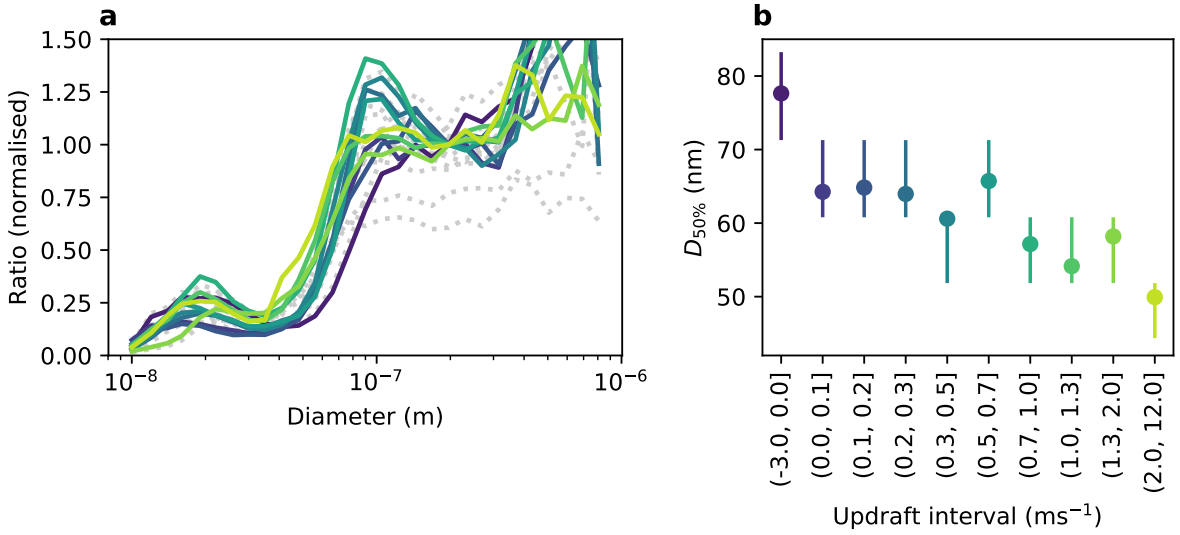

Figure 1: **$D_{50\%}$ dependence on updraft.** **a** Ratio of mean size distributions, i.e. cloud residual concentrations divided by total particle concentrations. The coloured ratio curves have been normalised to 1 at 200 nm. The grey dotted curves represent the position of the non-normalised ratio curves (Fig. 6 in main manuscript). **b** Interpolated $D_{50\%}$ for each updraft interval (i.e. the diameter at which the ratio in **a** is 0.5). The "error bars" represent the width of the diameter bin within which each interpolated value falls.

L.415: All observed updraft velocities are influenced by the orography independent of the amount of the updraft. Without the orography the updraft velocity would be different for an arctic cloud. Regarding in addition the point before, one would find an orographic influence on the D50%, which is again in contrast to the conclusion in the manuscript and needs to be further considered by the authors.

This is correct; however, updrafts close to 1 ms^{-1} are plausible for stratiform and stratocumulus clouds (and what physical forcing causes the updraft velocity is not really important). We have removed the sentence from the revised manuscript, but added the following where we discuss the $D_{50\%}$: "Updraft velocities in marine stratiform clouds are typically below 1 ms^{-1} (Zheng et al., 2016) and hence higher updraft velocities could be indicative of local orographic effects and may not be representative for Arctic clouds in other areas. Excluding the bins with updraft > 1 ms^{-1} gives a $D_{50\%}$ range of 58–78 nm (Fig. S8)". See also the response to the comments above and below with regards to the $D_{50\%}$.

L.418: Does it mean the enrichment factor is uncertain and only assumed? This should be clarified and made clear in the manuscript.

The enrichment factor is not only assumed; it is calculated based on the flows and geometry of the inlet (this is already mentioned in Sect. 2.2.2). There are technically always small uncertainties in e.g. flow measurements, which is why "and enrichment factor" was included in this sentence to begin with, but in the grand scheme of things such uncertainties are likely to be negligible. We realise now that mentioning them only adds confusion, so to address this comment we simply removed the phrase "and enrichment factor" from the sentence in question.

L.425-426: This is exactly what is brought up in the last two points and emphasizes that the GCVI sampling efficiency has to be determined for individual cloud events.

Following comment 5 above, we have implemented an individual correction factor for each cloud residual size distribution scan. This, however, does not significantly change the behaviour this comment refers to. The Shingler et al. (2012) transmission efficiency we use for the corrections can only account for what happens when the cloud particles enter the inlet/wind tunnel, but it could be that high updrafts or wind speeds prevent some cloud particles from actually entering the wind tunnel, and this is the kind of sampling issue that is meant in the text. We have added a parenthesis "(e.g. if the winds make it more difficult for the cloud particles to enter the wind tunnel)" to clarify this.

Fig.7: The first temperature bin from -8 to -21 C is much too broad. In this temperature range the cloud phase could change from supercooled to totally ice including all stages of mixed-phase conditions. The reason for this broad range is most likely a statistical one. It is not known whether most included cloud events are closer to -8 or -21 C and dominate the presentation and interpretation in and of Fig.7b and 7c. This should be presented in a more differentiated way.

It was indeed chosen for statistical reasons; however, we agree with the reviewer and have therefore split this bin to mitigate the issue. As can be seen in Fig. 2 below, the two "parts" of the original bin show very similar behaviour. We chose to put the additional bin boundary at -12°C because of the distribution of data points in the original bin (see Fig. 3 below).

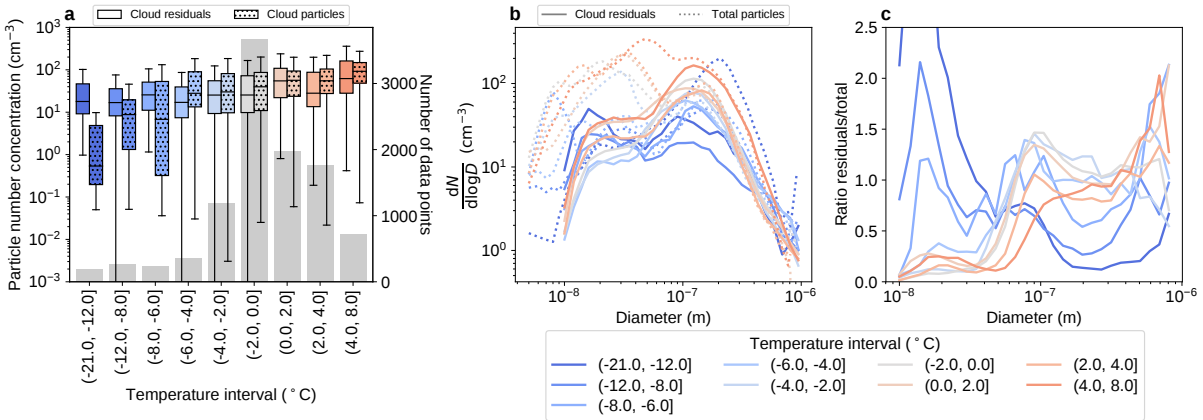

Figure 2: Updated version of Fig. 7 in main manuscript.

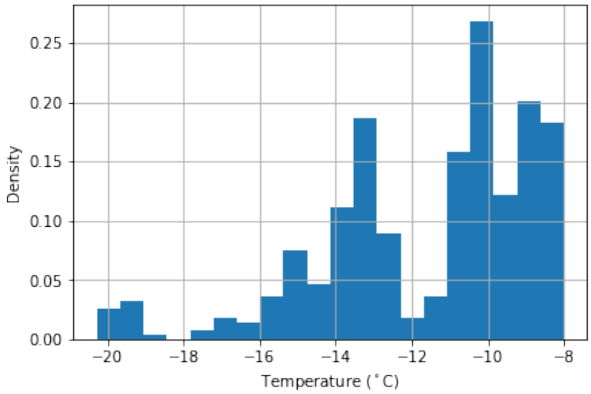

Figure 3: Histogram showing the distribution of temperatures in the original first bin (-8 to -21°C).

L.446-452: Many of these speculations coming up by the pure statistical approach of the data, and could be checked by analysing such clouds individually, which unfortunately is totally refused in this work, although it would significantly improve the investigation.

This paragraph just describes Figure 7b and the underlying hypotheses are later discussed in more (balanced) detail. Nevertheless, we have added two case studies to the revised manuscript that describe the strengths and weaknesses of our approach on individual cases.

L.502: The "some ice processes" has to be described in detail in the text and maybe "included" is a better mode of expression than "involved" here.

We have changed the sentence to "Therefore, the differences between our observed concentrations could either be because the actual ambient supersaturations are lower than what is used in CCN counters, or because the data in Fig. 7b still include some ice processes that can influence the droplet concentration (e.g. Wegener–Bergeron–Findeisen) while CCN counters only consider liquid droplet activation."

L. 537-542: This argumentation is straight forward and casts doubts on the conclusions drawn in section 3.2.1., like they are already expressed further up. The authors should seriously think about to change their data treatment in the mentioned section.

We fully agree with this, and changed this sentence so it just says "A relationship between updraft velocity and $D_{50\%}$ was also seen in Sect. 3.2.1" since that is indeed what we found when we tried the approach suggested by the reviewer of normalising the activation ratios to 1 (see Fig. 1 and responses to comments 5 and L.425–426).

Fig.12: In the lower row the peaks and the decrease in the ratios to 0.5 or lower for larger diameters look very suspicious. What are the reasons for these shapes. It is still in a size region where low counting statistics does not play a role. An explanation in the manuscript is definitely needed.

In section 3.2.1 of the manuscript, it is written "In the mid-size range, the ratio fluctuations could be the result of small uncertainties in sizing, concentration and losses of the two DMPS systems, causing the size modes to not be perfectly aligned". If the cloud residual size distribu-

tion is shifted to the left with respect to the whole-air particle size distribution (due to sizing uncertainties or perhaps evaporation of volatile compounds from the residuals), this could cause the observed "wiggly" behaviour of the ratios. We did not repeat this reasoning in the context of Fig. 12, but we agree that it would be good to do so. We have added the sentence "In the accumulation mode size range, the ratios show a wavy behaviour where, instead of levelling out around 1, they drop and then increase again. As stated in Sect. 3.2.1, this could be a result of the cloud residual and total particle size distributions not being perfectly aligned (e.g. due to uncertainties in sizing, losses, or perhaps evaporation of volatile material from the residuals)."

L.560-562: When looking at Fig. S8 this statement seems to be not correct. Here are residual size distributions visible that are very similar to cluster 1 but measured at temperatures above -4C, where clouds are mostly still liquid. The authors should comment on this point.

Cluster 1 is associated with much colder temperatures than the other clusters, this much is non-disputable. However, we agree that with the previously drawn boundary between warm and cold at -4°C, the sentence could be improved. We have modified the sentence so it reads "Cluster 1 occurs primarily during the winter months, and at considerably higher visibility and lower temperature and cloud particle concentrations than the other clusters (Fig. 9)".

Later on in the discussion, we conclude that cluster 1 is most likely significantly influenced by artefacts from snowflake shattering. While cloud particles may indeed mostly be liquid droplets at ambient temperatures above -4°C, those temperatures in no way preclude the occurrence of snowfall, so Fig. S8 is not inconsistent with the discussion of cluster 1. As mentioned earlier, the cluster analysis has been performed to avoid a simple discrimination by temperature, month or updraft but look specifically for the differences in (normalised) particle size distributions of the residuals.

L.562-565: The manner how the Cloudnet information is used here is a little bit misleading. Cloudnet only indicates the frequency of ice, mixed-phase and liquid clouds, but not the ratio of crystals to droplets in a mixed-phase cloud, which is mostly dominated by droplets in terms of number. Moreover, only clouds at temperatures higher than -21C were studied. At this temperature clouds are very rarely pure ice clouds. Thus, this text passage is not very convincing.

All clusters also have liquid cases included, so we do not talk about pure ice clouds here. We agree that the phrasing might not be clear, so we have clarified that we are talking about the ratio of ice to liquid cases and not the ratio of ice to liquid in an individual cloud. The sentences now reads: "The Cloudnet analysis shows that cluster 1 has by far the highest occurrence of cases with ice crystals compared to cases with liquid droplets (Figs. 10 and S11a). Interestingly, the ratio of ice to liquid cases decreases from cluster 1 to cluster 5, which is consistent with the activation ratios in Fig. 11 which appear more like classical Köhler activation (of homogeneously mixed particles) when moving from cluster 1 to cluster 5."

L.569: The citation of Verheggen et al. (2007) is appropriate to document the WBF process but is inappropriate to explain cluster 1 at this place. Fig.3 in Verheggen et al. (2007) shows the same ratio as a function of particle diameter and those graphs steadily increase with diameter and show no maximum at 20 nm in contrast to Fig.12a. Thus, this reference must not be used in a wrong sense to justify the existence of cluster 1 size distributions in cold clouds found in another publication.

Within this paragraph we argue that the accumulation mode is depleted due to the WBF process

as also observed by Verheggen et al. (2007) and we disagree that we use the reference in a wrong sense here. However, to clarify to the reader that this does not explain the shape of cluster 1, we modified the sentence: "This might be an explanation for the missing accumulation mode in cluster 1; however, it does not explain the peak activated fraction we observe around 20 nm (which was not observed in Verheggen et al. (2007))".

L.572-573: The authors need to specify the "cold temperatures" exactly. According to the presented data, the temperatures are in a range that it is very unlikely to have pure ice clouds. Moreover, the cloud particle concentrations are in the several cm-3 range according to Fig.S8, which is very high for a pure ice cloud. Again, an appropriate sensor to support this speculation is missing, so that this sentence and statement should be reworded or left out.

Here, the reviewer contradicts the comment L. 383–389. We have a group of data points where the FM-120 shows concentrations below 1 cm^{-3} at temperatures around -10°C or lower, which the reviewer himself says "must be more or less totally glaciated clouds" (see answer to that comment for some more on this topic). However, it is true that slightly less than half of cluster 1 contains such data points, so we have reworded the sentence to point out that pure ice clouds are only plausible in some of the cases: "Part of cluster 1 is associated with very low cloud particle concentrations ($< 1\,cm^{-3}$) and cold temperatures (down to -21°C), which may also be consistent with pure ice clouds."

As for Fig. S8, there are two things to point out. 1) The clusters and monthly data cannot be equated. Each month is a mixture of all clusters, which of course also affects the average number concentrations. 2) The concentrations are from the GCVI data, not the FM-120. If there are artefacts or sampling issues involved, those concentrations may not agree. We have added the FM-120 concentrations to the figure as well to highlight this.

L. 575-578: Again, only half of the truth is told here. Namely, the similar shape of the residual particle size distribution to presented ones in two other publications. But it is not taken into account that the Seifert et al. (2003) study is definitely carried out in pure ice clouds and that Mertes et al. (2007) used indeed a phase separating CVI inlet, which in addition was especially designed to avoid droplet and ice particles shattering, which is not the case in this study. What is additionally concealed is the fact, that the simultaneously measured background aerosol looks completely different to the one in this study. Consequently the "ratio" looks totally different compared to the ones in this study which make up cluster 1 (Fig.7c). The interpretation of the residual size distribution should always include the ambient background particle size distribution, which proofs in Mertes et al. (2007) and e.g. also in Kupizewski et al. (2016) [JGR] that small particles does not play a role in mixed-phase cloud formation. These facts need to be included here, in order to prevent that the reader gets a wrong impression by the stated "similarity" of the present study to the former ones.

The differences in measurement techniques and cloud types of the above mentioned studies are clearly mentioned in the sentence this comment refers to ("[...] previously been observed for ice particles in mixed-phase clouds measured with an Ice-CVI (Mertes et al., 2007), as well as for cirrus clouds using an airborne CVI (Seifert et al., 2003)"), and in addition repeated in the next sentence ("Although those studies used different techniques and sampled different cloud types [...]"). However, we agree that we should also mention the difference in the background aerosol size distribution and the activation ratios, so we have added the following sentence: "It should, however, be noted that the total aerosol size distributions in the aforementioned studies look

different than in the present study, and consequently their activated fractions do not show the same behaviour as our cluster 1."

L. 595-597: I do not understand the first sentence and therefore in addition not the message about decreased influence from snow. With the second sentence the authors left the reader alone with the decision to refer the observation to a measurement artefact or to a real cloud process, which is a not goal oriented approach to my mind. So here it would be desirable to make the statements more clearer and more determined.

We have tried to clarify the sentences so the text now reads: "However, cloud residual size distributions with modal diameters similar to those of clusters 1 and 2 still appear in a cluster analysis where all the data outside the 10:1 and 1:10 lines in Fig. 2b are excluded (Fig S13a; note, results do not change if we are even stricter, i.e. within 1:2 and 2:1). This stricter cluster analysis also shows a cloud particle mode for the small cloud residual cluster (Fig S13a), as opposed to the flat cloud particle size distribution for cluster 1 in Fig. 8, indicating a decreased relative influence from snow in the stricter analysis. The fact that cloud residual size distributions similar to cluster 1 still appear in Fig S13 suggests that while ice crystal shattering is certainly a possibility, it is not necessarily the only explanation for the shape of the size distributions we observe."

As for the "goal oriented approach", the issue is that we cannot definitively say that the small residuals are only a result of artefacts, just as we cannot say they are entirely caused by real processes. Artefacts most likely do play a role in cluster 1, which we also conclude in the manuscript, but because there are cases when artefacts are not the obvious cause (e.g. cluster 1 or 2 when the agreement with the fog monitor is good) we also think it is important to discuss possible physical explanations (like the secondary ice processes, although we know the reviewer is not a fan of this particular hypothesis). The possibility that artefacts and real processes could produce similar signals is also something that is worth highlighting, since it is an important consideration for others planning to carry out GCVI measurements.

L.615: What is meant by the nuclei here? An INP and/or CCN or something else? It is hard to believe that an INP will be fragmented by an ice particle shattering during ice-ice collisions. Are there any indications that INPs undergo fragmentation? It is much more likely that small residuals stem from CCN matter build in the ice lattice. Since ice particles in mixed-phase clouds are mainly formed by droplet freezing (immersion and contact freezing), the CCN matter should be still in the ice particles and condense to small particles during the ice sublimation in the CVI in the absence of the INP. However, I would not call it "fragments of CCN", since it is not a mechanical fragmentation.

The process described by the reviewer is what we are hypothesising could cause the small size of the residuals. We concede that we did not explain it adequately, and have clarified the text and also replaced the word "fragments" with "remnants". The text now reads: "In ice crystals formed by droplet freezing, solute material from CCN or scavenged particles could be built into the crystal structure, and that material could then be distributed across the splinters when secondary ice formation happens. In other words, the cloud residuals we measure may be remnants of CCN and/or scavenged particles, which would explain their small size."

L.618-619: So, the absence of small cloud particles is a clear indication for an artefact sampling. But here it is presented as a marginal contribution with respect to the observation of secondary ice. This is a wrong formulation, since the existence of secondary ice is not proven

and thus also not the connection to the observation of small residual particles. Therefore, the original statement here needs to be rebutted by rewording this sentence.

Here, we are not sure we understand what the reviewer means.

The argumentation of lines 609–629 is basically: Secondary ice could be an explanation for the small residuals. If secondary ice formation happened, it should also be seen by the fog monitor (unless the ice crystals are undersampled by the fog monitor); however, the cloud residual and cloud particle concentrations often do not agree in cluster 1. Therefore, secondary ice cannot be the only explanation for the small residuals. Cluster 1 is significantly affected by snow and ice sampling, and most likely includes artefacts.

This is not different from what the reviewer says, so we have not modified the text.

L. 628-629: It is not clear, why small cloud particles must be ice fragments from secondary ice processes only. The authors themselves refer to the Wegeron-Bergeron-Findeisen process, where droplets lose water for the growth of ice particles and therefore getting smaller. So small particles in mixed-phase clouds could also be evaporating droplets. The authors should also take this possibility into account.

We apologise that the sentence was unclear, the particles referred to are the cloud residuals, not the cloud particles. We replaced "particles" with "cloud residuals" to make it clearer. The intended message of the sentence is that small cloud residuals such as those in cluster 1, could either be caused by artefacts or potentially by secondary ice processes like the one the reviewer describes in comment L.615 above.

L. 641-643: To support the interpretation of the cluster 2 size distribution, it would be good to estimate the maximal supersaturation in the cases this cluster was observed. This should be possible with the available measurements of temperature, cloud base height and vertical wind speed and would reveal if particles with a diameter of 30 nm (mode diameter of cluster 2) can be activated and how soluble they have to be. This is done in the cited reference Schwarzenboeck et al. (2000) as well as in the accompanying model study by Grmy et al. (2000) [Tellus, 52B, 959-979] to evaluate the observation of small residuals in warm clouds and would be a profit here as well.

We believe that this request is out of the scope for this work. These calculations are also not as straight-forward as the reviewer might think, due to the fact that the cloud radar is located at the village of Ny-Ålesund (so approx. 2 km away) and the cloud base height is also often difficult to retrieve due to the low altitude of the clouds. But most importantly, there is a number of theoretical and experimental studies that have shown the possibility of Aitken mode particles to activate. Recently, Bulatovic et al. (2021) have used more sophisticated large-eddy simulations and have shown again that Aitken-mode particles play a crucial role for Arctic clouds. We have added this new reference to the revised manuscript.

L. 672: I may have overlooked, that the term "cloud nuclei" was defined, but I would prefer still name it CCN and INP.

We replaced "cloud nuclei" with "CCN" here.

L. 676: To activate 20 nm particles is even more difficult as 30 nm particles. With this statement, a calculation of the max. super-saturation is indeed needed to see if this possible at all and if yes, for which type of aerosol particles.

Other modelling studies have shown the possibility to activate particles at theses sizes and we see no need to repeat their work in calculating the max. supersaturations. For example, Bulatovic et al. (2021) recently showed in a comprehensive study the potential importance of Aitken mode particles $\sim 25 - 80$ nm for stratiform Arctic mixed-phase clouds (we have added this reference to the revised mansucript). Of course, with respect to standard Köhler theory, it is much more difficult to activate a 20 nm particle compared to a 30 nm particle. For a 20-nm ammonium sulphate particle one would expect a critical saturation ratio of 2.41 to 1.98% at temperatures between 250 and 280 Kelvin. For more hygroscipic particles like sodium chloride, the critical ratio would decrease to 1.54 to 1.29 %. The corresponding values decrease substantially for 30 nm (approx. main mode of cluster 2) to values between 0.70 and 1.29. These values are high but it is not impossible to activate those small particles. In addition, we already mention here and throughout the manuscript that potential artefacts can not be fully ruled out. Since our mode is more around 30 nm, we added this number to the respective sentence.

2 Anonymous Referee #3

The revised manuscript "A long-term study of cloud residuals from low-level Arctic clouds" has largely addressed most of the comments of the reviewers. However, a few comments remain not be thoroughly addressed. And I have some additional comments. I recommend the publication of this manuscript in ACP if these comments can be addressed.

1. Both reviewers mentioned making comparison for the particle number distribution in cloud and outside cloud to ensure the measurement quality from whole-air inlet (Reviewer 1 comment 3 and 7, Reviewer 2 minor general comment 6). Cloud particles may also experience possible additional losses in the whole-air inlet too due to e.g. the different size distribution compared with ambient aerosol particles. But the authors decided to not follow the comments because the authors argue that "appearance or termination of the cloud event can also be caused by a change in air mass with differences in aerosol properties." In my opinion, the information on the particle loss of whole-air inlet for cloud droplet sampling is necessary because if there were significant difference, it would affect the ratio of cloud residuals to total particle concentrations shown in Fig. 6 and Fig. 7. Although the authors showed a new comparison of cloud residual concentration vs. total particle concentration in accumulation mode, this does not necessarily mean that total particle concentration has no artefacts/free of loss and of ensured quality.

As will be described in the following comment, our inlet follows the ACTRIS recommendations for sampling in extreme environments and it generally compares very well with other instrumentation at different inlets at the station (see comment below). But most importantly, we would like to bring the argument forward that the comparison of the accumulation mode of whole-air and CVI inlet gives the exact same factor (0.46, see revised manuscript) as the expected CVI sampling efficiency factor determined from the ambient fog monitor measurements (see Fig. 3 in revised manuscript), which is almost too good to be true. As such, we see no reason to suspect major losses within the whole-air inlet sampling during cloudy periods. We have also added two case studies to the revised manuscript, where the periods before and after the cloud event are included.

2. A related comment to the comment 1 is that the description of the whole-air inlet is lack, especially the important role of the data for this study. One cannot expect readers to follow the setup just stating "fulfils the World Meteorological Organization guideline". For example, how is the inlet heated, and to what temperature? Has this setup been validated previously regarding the particle loss? If so, references could be cited. More details would be helpful here.

The inlet was built according the guidelines of ACTRIS (https://www.actris.eu/) and the WCCAP (World Calibration Centre for Aerosol Physics) in close consultation with Institute of Tropospheric Research (TROPOS), Germany. A detailed description can be found here (https://www.wmo-gaw-wcc-aerosol-physics.org/recommen-dations.html, document: "WCCAP recommendation for aerosol inlets and sampling tubes"). To prevent freezing, the inlet is heated and kept at temperatures between 5 and 10 °C. As already mentioned in the manuscript, the inlet has similar characteristics as the inlet at Jungfraujoch (Switzerland) as described in Weingartner et al. (1999). The inlet has not been experimentally validated for particle losses. In general, there is an overall excellent agreement with other instrumentation on other independent inlets of the observatory that we have tested in other work (e.g. to the FIDAS instrument on the terrace) but there is no current reference to it. To support this fact for this respective study, we like to bring forward the argument that the two independent methods of determining the CVI sampling efficiency (accumulation mode comparison using CVI inlet/whole-air-inlet and derivation via the ambient cloud particle concentration using the CVI inlet/fog monitor) have given the exact same value of 0.46. This demonstrates, at least for the accumulation mode, an excellent performance of the whole-air inlet. For clarification, we have added to following to the revised manuscript:
"The inlet follows the guidelines for whole-air inlets for extreme environments given by the World Calibration Centre for Aerosol Physics (WCCAP) at the Leibniz Institute for Tropospheric Research, Germany (https://www.wmo-gaw-wcc-aerosol-physics.org/recommen-dations.html). It ..."
and
"...to a temperature of around 5-10°C to prevent freezing..."

3. In the new structure, for Section "3.2.1 Warm clouds" and Fig. 6, are all these data for temperature ¡ 0 C? If so, it would helpful to clarify explicitly in both the text of 3.2.1 and caption of Fig. 6.

Yes, this is correct. We have added the sentence "This subsection only deals with data collected at temperatures > 0°C" to the introduction of 3.2.1 and the sentence "Only data collected at temperatures > 0°C are shown in this figure" to the caption of Fig. 6 to clarify this.

4. Section 3.2.2 heading "Cold clouds" seems not to be consistent with the scope of discussion that it covers. It mainly discusses the influence of temperature rather than cold clouds. Maybe a different heading would work better.

We chose those headings to mirror the headings in section 3.1, but we agree that it is perhaps not suitable in this case. We have renamed 3.2.2 "Influence of temperature", and to be consistent we also renamed 3.2.1 "Influence of updraft" (while still specifying that 3.2.1 only deals with warm clouds, see answer to comment above).

5. Some responses to reviewers should be incorporated to the revised manuscript as they will be helpful to other readers. For example, the response to the comment D (Pg 17 of the authors

response file), comment "L. 158-159" (Pg 23 of the authors response), comment "L121-122" (Pg22), comment.

We agree. In the revised manuscript, we have added the information from commend D. In the conclusion, we modified a sentence (new part in italics) "To study ice and liquid cloud particles separately, it would also be desirable to deploy ice-selective inlets (e.g., Mertes et al., 2007; Kupiszewski et al., 2015; Hiranuma et al., 2016) at Zeppelin Observatory in the future; *however, long-term deployment and potential artefacts remain a challenge*", see also the response to comment 4 by Reviewer#2. The link to the work by Verheggen et al. (2007) was already implemented in the last revised version.

Concerning comment "L. 158-159", we have added "With this set-up two total CPCs are available and the second CPC is used as back-up and quality assurance." as well as "For the number concentrations shown in the manuscript, we used the integrated and loss corrected particle number size distributions. However, when comparing the cloud residual number concentrations to the cloud particle concentrations, the total CPC (behind the GCVI) was used." to the method section.

Concerning comment "L121-122", most of the information was already included in the revised manuscript but we added/modified the following sentence: "The likelihood of shattering can be estimated by the non-dimensional Weber number, where fragments are expected to be produced under conditions with Weber numbers between 10 and 12 (Twohy et al., 2003). In the GCVI, these conditions occur at 100 ms^{-1} air speed and droplet diameters between 70 and 100 μm (F. Brechtel, pers. comm., Oct. 2020)."

6. Some "new" statements are introduced in the "conclusions" part while how they are drawn is not straight and completely clear in the main text. For example, L663, "The cluster analysis of cloud residual size distributions showed a D50% dependence on updraft for liquid clouds (clusters 35)." This finding is not explicitly drawn, although the authors show that D50% depends on updraft velocity and updraft velocity of clusters 35 are different. A direct way to draw such a finding would be a figure showing the D50% dependence on updraft for data belonging to clusters 35. L666-667, the finding "A clear relationship between a decreasing total particle number concentration and a decrease in D50% was also observed" is also not directly drawn in the main text. In L537-538, a figure of D50% vs. particle number concentration and updraft velocity is needed as the current presentation of the trend e.g. D50% values is hard to follow. L668-670, it is not clear which data and discussion these statements are based on since the data were not discussed by April-October or November-March.

We agree with this comment. Concerning the $D_{50\%}$, we have added a new figure in the SI (see Fig. 1 above) where we explicitly show the updraft dependence of the activation diameters for warm clouds. As for the dependence on number concentration, we saw a correlation between $D_{50\%}$ and temperature in Fig.7c, and temperature, in turn, correlated with number concentration in Fig.7a. We have changed the conclusions and used the word "inferred" instead since the relationship was not explicitly shown. As for clusters 3–5, the conclusions were also drawn more implicitly in the main text and we modified the conclusions to better reflect this. In summary, we have changed the paragraph in the conclusions to: "For pure liquid clouds (T> 0°C), we observed activation diameters ($D_{50\%}$) in the range of 58–78 nm (for updraft velocities below 1 ms^{-1}), where smaller activation diameters were associated with higher updraft velocities. A relationship between a decreasing total particle number concentration and a decrease in $D_{50\%}$ could also be inferred from the cloud residual size distributions binned by temperature. Both

a change in updraft velocity and a change in particle number concentration can affect the supersaturation, but we cannot clearly disentangle the influence of these parameters. The cluster analysis of cloud residual size distributions for liquid clouds (clusters 3–5) also showed that smaller cloud residuals were associated with higher updraft velocities and lower particle number concentrations."

Concerning the last part of the comment, the periods are based on the cluster occurrence plot (Fig.9b). It is true that the main text was more general (i.e. winter, summer, etc.). We have modified this part of the conclusions to hopefully make it more clear (new parts in italics): "*From late spring to early autumn*, the cloud residual size distributions at Zeppelin Observatory are dominated for most of the time by the accumulation mode with clouds consisting mostly of liquid droplets *(clusters 3–5)*. In *late autumn to early spring*, we found, in relative terms, a significant contribution of Aitken mode particles to the cloud residual number concentration *(clusters 1 and 2)*."

7. The abstract has not fully reflected the changes in the focus of the revised manuscript as much of the findings in the main text and conclusion are missed in the abstract.

We agree. We have edited and extended the abstract so it now reads (changes are marked in italics): "To constrain uncertainties in radiative forcings associated with aerosol–cloud interactions, improved understanding of Arctic cloud formation is required, yet long-term measurements of the relevant cloud and aerosol properties remain sparse. We present the first long-term study of cloud residuals, i.e. particles that were involved in cloud formation and cloud processes, in Arctic low-level clouds measured at Zeppelin Observatory, Svalbard. To continuously sample cloud droplets and ice crystals and separate them from non-activated aerosol, a ground-based counter-flow virtual impactor inlet system (GCVI) was used. A detailed evaluation of the GCVI measurements, using concurrent cloud particle size distributions, meteorological parameters, and aerosol measurements, is presented for both warm and cold clouds, *and the potential contribution of sampling artefacts is discussed in detail. We find an excellent agreement of the GCVI sampling efficiency of liquid clouds using two independent approaches.* The two-year data set of cloud residual size distributions and number concentrations reveals that the cloud residuals follow the typical seasonal cycle of Arctic aerosol, with a maximum concentration in spring and summer and a minimum concentration in the late *autumn* and winter months. We observed *average* activation diameters in the range 58–78 nm *for updraft velocities below 1 ms^{-1}. A cluster analysis also revealed cloud residual size distributions that were dominated by Aitken mode particles down to around 20–30 nm.* During the winter months, some of these small particles may be the result of ice, snow or ice crystal shattering artefacts in the GCVI inlet; however, cloud residuals down to 20 nm in size were also observed during conditions when artefacts are less likely."

Minor comments:

1. L523 and Fig. S9, why different clusters (2-5) show different slopes? Shouldnt it be that all cluster have similar slopes if the transmission efficiency of GCVI (size-dependent if I understand correctly) are correctly applied and the GVI is free of artefacts?

The GCVI transmission efficiency depends on the size of the cloud particles, and so the integrated transmission efficiency is a function of the cloud particle number size distribution. The agreement between the cloud residual and cloud particle concentrations after the transmission efficiency

correction has been applied depends on the validity of the correction for each individual case as well as artefacts or sampling issues in the GCVI and/or FM-120. It is true that, in an ideal world, the slopes would be the same. However, as discussed in the manuscript, because there are multiple instruments and corrections involved, and they each have their own uncertainties, it is not unexpected that the agreement is not perfect.

2. The caption of Fig. 2 can still be misleading if one reads separately from the main text. I suggest rewriting "cloud particle number concentrations derived from the FM-120 fog monitor measurements (red)" as "corresponding cloud particle number concentrations derived from the FM-120 fog monitor measurements and transmission efficiency of GCVI".

We agree and have reworded the phrase, it now says "corresponding cloud particle number concentrations derived from the FM-120 fog monitor measurements and the transmission efficiency of the GCVI inlet (red)".

3. L445, Fig. 6 or Fig. 7?

This should indeed be Fig. 7, thank you for noticing the mistake.

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