Reply to reviewers of the manuscript "The role of nanoparticles in Arctic cloud formation"

Karlsson et al.

December 8, 2020

We thank both reviewers for their very detailed and constructive comments. Both reviewers had concerns about the potential influence of measurement artefacts and both suggested that we should restructure our manuscript with a more pronounced focus on the actual cloud phase (liquid vs. mixed-phase). Overall, we agree with these major remarks and have therefore substantially restructured our manuscript (mainly the result and discussion part) and added additional analysis and text. In summary, these major changes include:

1. We have separated the analysis and the presentation of the results between liquid and mixed-phase clouds. This separation is done in the result part for both (a) the GCVI sampling efficiency and (b) the presentation of the two year data set of cloud residual size distribution measurements. Figures and text were adapted accordingly.

2. We now give detailed information on the GCVI and DMPS data treatment (new subsection in the method part) and provide further technical specifications in the main manuscript as well as in the supplementary information (SI).

3. We have added additional analysis with regard to the CVI sampling efficiency by comparing the accumulation mode concentrations of residual and total particle size distribution for liquid clouds, as suggested by the reviewer. The result of a sampling efficiency of around 0.5 is similar to the value determined from independent cloud particle measurements, which further supports our findings and the overall reliability of our data.

4. We have toned down the interpretation and implications with regards to the sub-100nm-particles, which was also needed after the restructuring and addition of new analysis. We now give more weight to the entire set of observations. We have also changed the title of our manuscript to a more general title ("A long-term study of cloud residuals from low-level Arctic clouds") describing the actual research performed.

5. Related to the point above, we are now more cautious in our conclusions with regards to the influence of sampling artefacts as well. Further discussions among the coauthors, sparked by comments from both reviewers, have led us to believe that a small amount of the Aitken mode cloud residuals in the winter may be the result of ice or snowflake shattering artefacts. Additional analysis and a more balanced discussion about this and other potential causes has been added in the revised manuscript.

Reviewer 2 insisted on entirely removing the data with the potential influence of mixed-phase clouds. Although the reviewer suggests that these data points are suspicious, we believe we

have shown that there is still a lot that can be learned from them. As outlined above, we have chosen to present the results for liquid and mixed-phase clouds separately instead of discarding any data. We believe that the new structure makes it clear to the reader which data points are likely to be affected by artefacts and which are not, and that the toning down of the conclusions relating to those data points now makes the paper well balanced, thus resolving the issues raised by the reviewer. As suggested by both reviewers, we tested the approach of excluding data where no agreement between the residual and cloud particle number concentration was found. Fortunately, the overall results did not change, which gave further confidence in the validity of our results.

We see no clear evidence of artefacts originating from the sampling of ice crystals inside inside the CVI but we do discuss the potential break-up of ice crystals before the CVI (artefacts generated within the wind tunnel) or in the atmosphere (secondary ice). By using the cluster analysis, we directly assess the contribution of residual size distributions that do not follow the expected classical behaviour for liquid droplet activation and by doing so, we deliberately investigate the overall contribution, the temporal evolution and the trustworthiness of those ice-influenced size distributions. We would also like to emphasise that similar cloud residual size distributions have been observed before using similar techniques (see e.g., Seifert et al., 2003, although for cirrus clouds) or (Mertes et al., 2007, using an ice-CVI), thus there is no reason to fully remove this data. One important addition to the revised manuscript relates to the findings of Verheggen et al. (2007), a study on mixed-phase clouds at the high-alpine site Jungfraujoch. Although Verheggen et al. (2007) used a slightly different inlet system set-up, the temperature-dependence of their activation ratios agrees surprisingly well with our data from the Arctic, giving further confidence in the overall validity of our work.

Further, we would like to stress that the same type of CVI has been previously thoroughly evaluated (Shingler et al., 2012). The CVI has been used extensively by other research groups on various aircraft campaigns (e.g., Modini et al., 2015; Sanchez et al., 2016; Hossein Mardi et al., 2019). Since the CVI was installed within a wind tunnel, we added information on how its performance within the wind tunnel (GCVI) was evaluated by the manufacturer. In addition, we would like to mention that the same GCVI has been successfully deployed within short-term campaigns by our and other research groups and, like for the aircraft studies, no major artefact production has been observed within the sampling line of the CVI (Zhang et al., 2017; Lin et al., 2019a,b; Graham et al., 2020; Baccarini et al., 2020).

Summarising, we are convinced that by adding additional analysis, information and discussion and by restructuring the result section, our manuscript has significantly improved in quality. We thank the reviewers again for their great effort and patience.

We will provide more detailed replies to their comments below. Our comments are given in blue, new text within the revised manuscript is given in light blue. Because the revisions are so substantial, we have not been able to include every single minor change in this document. We have included the main ones in response to the relevant comments, and refer to the diff document for the complete set of changes. We have not included the new figures within this reply letter but rather refer to the revised manuscript and the SI.

1 Anonymous Referee #1

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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.

We thank reviewer 1 for their detailed and helpful comments. We have followed the advice to start the discussion of our findings with the data of liquid clouds and finishing the discussion for mixed-phase clouds. We emphasise that artefacts cannot be ruled out, and have expanded the discussion (including additional figures) to try to quantify when artefacts are most likely, when they are less likely, and what bearing they have on the results. Further details are given below. This is the new structure of the manuscript (new headings/headings with changed names are in cyan instead of blue):

1.1 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.

We fully agree with this comment. We have restructured our manuscript and give more weight to the overall findings for all cloud types (liquid vs. mixed-phase/ice). We have expanded the discussion and include the possibility that clusters 1 and 2 are affected by artefacts, clearly stating that this cannot be ruled out. However, in contrast to what the reviewer suggests, we believe Cluster 2 is influenced to a much lesser extent than Cluster 1.

Further discussions among the coauthors, sparked by comments from both reviewers, have led us to believe that at least part of the Aitken mode cloud residuals in the winter may be the result of snowflake shattering artefacts, and this discussion has been added in the updated version of the manuscript. We suspect snow because particles need to reach a certain size for shattering to be likely, and this size ($\sim 70\,\mu$m) is far larger than the main mode seen in the FM-120. We

have added this in Sect. 3.1.3: "At air speeds of 100 ms^{-1}, the critical diameter above which droplets may shatter into fragments is $\sim$ 76 μm (Twohy et al., 2003). Precipitating particles can exceed this size, and could thus produce fragments that are sampled if they are larger than the aerodynamic cut-size of the GCVI. However, it is likely that many of the fragments would not be aligned with the streamlines and therefore would not enter the GCVI."
There may be a few cloud droplets or ice crystals that are large enough to shatter but, as both reviewers have pointed out, such particles should be far outnumbered by the main droplet mode. Thus, they would have to be very large indeed to shatter into enough fragments to significantly affect the measured cloud residual distributions (bearing in mind that fragments still need to exceed the GCVI cut-size and be aligned with the streamlines in order to be sampled). We have also modified/added this in Sect. 3.1.3: "Droplet or ice crystal shattering is another potential source of small particles. Shattering could either happen in the wind tunnel or after the stagnation plane within the CVI inlet, and this could also cause an overestimation of the cloud residual number concentration. If the particles were to shatter after the stagnation plane, this should be clearly seen as spikes in the cloud residual concentrations measured by the total CPC, and this was not observed. Regarding shattering in the wind tunnel, as stated above, the cloud particles need to exceed a certain size for this process to be likely. While this can happen when there is precipitation, it needs to be borne in mind that in most non-precipitating clouds, the concentration of large cloud particles is much lower than the total cloud particle concentration, and therefore particles that do shatter may need to far exceed the critical break-up diameter to produce enough fragments to significantly increase the measured cloud residual concentration (Twohy et al., 2003). If the concentration differences we observe are caused by shattering artefacts, then, the magnitude of the difference suggests that precipitating particles (e.g. snow) are a more likely cause than large cloud droplets or ice crystals."
Cluster 1, which is linked to cold temperatures and has no clear droplet mode, is therefore likely to be influenced by snow shattering artefacts. Cluster 2, on the other hand, has a clear droplet mode and has a temperature distribution that is not appreciably different from those of clusters 3–5. In addition, its shape is not so much flat as bimodal with two broad modes. Within the new Fig. 11 one can observe that the average residual number size distribution consists of two broad modes (roughly around 30 and 150 nm), which are also present in the concurrent total size distribution measured behind the whole-air inlet. However, it is possible that the influence of artefacts gives the size distribution a flatter appearance — when the cluster analysis is repeated without the most suspicious data points (new supplement Fig. S12) the bimodality can be seen more clearly, which supports this hypothesis. We have added a supplementary figure with the cloud residual vs cloud particle concentration comparison separated by cluster to help quantify the influence of artefacts/suspicious data, and have added this information to the discussion about the clusters as well: "Large crystals are expected to be more prone to shattering, and indeed clusters 1 and 2 are related to larger cloud particles than the other clusters (Fig. 9a). Cloudnet does not distinguish between cloud ice and precipitating ice, so we could also be dealing with snowflakes. The average cloud particle size distribution associated with cluster 1 is rather flat with no obvious mode (Fig. 9a), and it is also associated with very low cloud particle concentrations (Fig. 10d). This could just be noisy measurements in the fog monitor during snowfall, and would indicate that cluster 1 is influenced by snow. Cluster 2, on the other hand, has a clear cloud particle size mode, although at a larger diameter compared to cluster 3–5 (Fig. 9a), and the cloud residuals are therefore much less likely to stem solely from precipitation.

The median cloud residual concentration is slightly larger than the median total particle concentration for cluster 1 around 20 nm (Fig. 12a), which would suggest that there is a risk of crystal shattering artefacts. As shown above, Figure 2b revealed two groups of data, where one showed

a discrepancy between measured cloud residual and cloud particle concentrations as would be expected with this type of artefact. The same figure separated by cluster (Fig. S9) shows that this group of data is overrepresented in clusters 1 and 2, which speaks in favour of the crystal shattering hypothesis as well. 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 S12a; note, results do not change if we are even stricter, i.e. within 1:2 and 2:1). Also note that a cloud particle mode then appears for the small particle cluster (Fig S12a), indicating a decreased relative influence from snow. This 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."
After some paragraphs about secondary ice (see response to next comment), we then conclude the discussion of clusters 1 and 2 with the following: "In summary, it seems likely that cluster 1 is significantly influenced by snow and ice. It is difficult to say to what extent the signal is caused by crystal shattering artefacts as compared to other processes, but cold temperature outliers (cf. Sect. 3.1.3) make up roughly 45% of cluster 1 (cf. Fig. S9a). These data should be treated with caution, but there are some plausible physical explanations for the presence of small particles when the agreement with the fog monitor is better, e.g. secondary ice processes, yet further measurements would be needed to verify this. The possibility that such processes would show a signal similar to shattering artefacts is an important consideration when analysing GCVI data from ice or mixed-phase cloud conditions. However, from an aerosol activation perspective, it is irrelevant whether the snow or ice crystals shatter before or after they enter the inlet – in both cases, the resulting cloud residuals do not represent cloud nuclei.

Cluster 2 is also Aitken mode dominated, and occurs throughout the year (27 % of the time, or 13 % of the time if we only consider $T > 0°C$). Unlike cluster 1, cluster 2 is much less likely to be affected by snow artefacts. While the exact contribution is difficult to quantify, cold temperature outliers only make up about 13% of cluster 2 (cf. Fig. S9b), i.e. there is a significantly better agreement with the fog monitor than for cluster 1. Cluster 2 is also different from clusters 3–5 but, in contrast to cluster 1, it was observed more homogeneously throughout the year. In further contrast to cluster 1, the meteorological parameters related to cluster 2 are not distinctly different from those related to clusters 3–5 (cf. Fig. 10). This means that cluster 2 was also observed during sampling conditions when we can safely rule out the influence of mixed-phase clouds and ice crystals. Many of the caveats listed above related to cluster 1 thus do not apply to cluster 2 to the same extent. In addition, cluster 2 does not show the lack of accumulation mode particle activation that complicated the interpretation of cluster 1. Hence, the Aitken mode cloud residuals in cluster 2 very likely contain activated aerosol particles. Similar findings were reported in previous CVI measurements (Schwarzenboeck et al., 2000), although not in the Arctic. In the Arctic, activation of Aitken mode aerosol particles has been shown by indirect means and model studies (e.g., Leaitch et al., 2016; Koike et al., 2019; Korhonen et al., 2008). "
We hope that this, together with the overall refocusing of the manuscript that gives all the data more equal weight, has balanced the paper in a satisfactory way.

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.atmoschem-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.

We agree. We have added the caveats concerning the secondary ice hypothesis to the discussion of the cluster analysis. We have also expanded the discussion about artefacts; please refer to the previous comment for more details. As the reviewer correctly points out, residuals that are the result of scavenged particles, secondary ice processes or shattering artefacts cannot be said to be driving cloud formation since the particles were not acting as cloud nuclei. We have clarified this point in the discussion about the clusters as well: "At this point, it is important to point out that, even barring artefacts, a cloud residual does not necessarily respond directly to a CCN or INP. Cloud residuals can also be nuclei that have undergone processing inside the cloud (be that chemical or physical), and can contain material from e.g. riming or aerosol particles that have been scavenged by the cloud particles. Unfortunately, we have no way of distinguishing between these particle types, especially since the FM-120 cannot differentiate between cloud droplets and ice crystals. It could potentially be that the crystals we measure are the result of secondary ice formation processes (Field et al., 2016), which has been suggested in a model study to be important for Arctic stratocumulus clouds (Sotiropoulou et al., 2020). In other words, the cloud residuals we measure may be fragments of nuclei and/or scavenged particles, which would explain their small size.

Since secondary ice formation happens before the cloud particles enter our inlets, these particles should also be seen by the fog monitor. This is often not the case for cluster 1, as seen in Fig. S9, which means that secondary ice particles cannot be the only reason for the small residuals we observe (unless the ice crystals are undersampled by the fog monitor, see Sect. 3.1.3). Cloud observations at mountain-top stations such as Zeppelin Observatory may also be influenced by surface processes (e.g. blowing snow) that could increase the ice crystal concentrations (Beck et al., 2018), but this, too, should be seen by the fog monitor and as discussed earlier, no clear dependency on wind-speed has been observed. As stated earlier, it is not possible to translate the cloud residual data to CCN, INP, etc without further detailed information on cloud phase, structure and origin. The cloud phase is an important parameter and should as such be added in future studies. "

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 authors 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.

The total particle number size distributions in Fig. S7 were measured behind the whole-air inlet

during the cloud event (measured concurrently with the cloud residual data), so it comes from both cloud particles and interstitial particles. As suggested by the reviewer, we have moved an updated version of this figure to the main part of the manuscript. The appearance or termination of the cloud event can also be caused by a change in air mass with differences in aerosol properties. As such, we refrain from an additional analysis of particle size distributions before and after the cloud event (cloud residual and whole-air) since we believe that the two-year (concurrent) dataset stands for itself.

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.

We agree that the terms "cloud residual", "CCN", and "INP" were (unintentionally) somewhat muddled in the previous version of the manuscript. We have gone through the manuscript and done our best to correct this issue, as well as adding clarifying statements (see response to comment 2 above).

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.

We have worked to fix this issue (see response to previous comment). We also believe that the new subdivision of the manuscript where cold and warm clouds are treated separately helps to avoid any confusion.

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.

We agree with this comment and have done the following improvements:

- We present warm (non-suspicious) and cold (more suspicious) data separately, and also

derive correction factors (from the fog monitor comparison) separately for warm and cold clouds (new transmission efficiency figures Figs. 2–4).

- We have added a supplementary figure (Fig. S9) showing the comparison between cloud particles and cloud residuals separately for each cluster, to help quantify the influence of suspicious data/potential artefacts on each cluster.
- We have added a supplementary figure with a new cluster analysis without the most suspicious data (Fig. S12), to show that the results do not change.
- We removed this part of the text.

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-chemphys.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?

As mentioned above, the appearance and disappearance of clouds can also coincide with a change in air mass. As such we refrain from performing extra analysis in comparing size distributions before and after cloud events. Most importantly, as now shown in the revised manuscript, the comparison of accumulation mode concentration measured behind the GCVI and the whole-air (=total) inlet, showed a remarkably good agreement (factor around 2 for liquid clouds) with the comparison of cloud residual concentration and cloud particle concentration (as measured by the FM-120). This clearly shows that the whole-air inlet at Zeppelin Observatory (which follows the ACTRIS guidelines for aerosol sampling in high-altitude/cold environments) is capable of sampling the entire population of ambient aerosol and cloud particles. In order to be consistent with the new structure of our result section, we have replaced the previous Figure S8 by a new figure showing the monthly averaged cloud residual size distributions (separated by all, warm and cold cloud cases). For this figure (Fig. S8 in the supplement) we decided to only show the cloud residual size distribution due to the larger data coverage (see Tab. S1).

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 – 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?

We looked at various cluster metrics to determine the optimal number of clusters, but the results were inconclusive. Some metrics suggested 2 clusters (e.g. silhouette scores), some suggested 3 clusters (e.g. elbow method / sum of squared errors), while others suggested as many as 17 clusters as the optimum (Davies-Bouldin index). For this reason we decided to test and to show the result for a number of clusters of up to 6 clusters (see Fig. S3 in the SI). It was our intention to find the optimal number of clusters that would carve out the characteristic cluster

with a domination of Aitken mode particles (here cluster 1). We used physical reasoning to find the optimal number of clusters, and by looking at e.g. Fig. 10 (in the revised manuscript) it is clear that cluster 1 is physically distinct from cluster 2. Increasing the number of clusters to more than 5 only lead to an additional split of the accumulation mode dominated residual size distributions (see Fig. S3). Previous studies using cluster analysis on particle size distributions usually use many more clusters (e.g. Beddows et al., 2009), in addition to own judgements in combining the "over-clustered" size distributions to fewer clusters later on (e.g. Dall'Osto et al., 2017; Tunved and Ström, 2019).

The total particle data was then grouped based on the retrieved clusters. Interestingly, if we remove the data points with large disagreement in the FM-120 vs GCVI comparison (points outside the 10:1 and 1:10 lines; see Fig. 2b), similar clusters appear in the cluster analysis (new supplementary Fig. S12) which gives further confidence in the overall results.

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.

We agree and have removed this part of the conclusions. Based on the all comments and the revisions done, we have completely rewritten the conclusions, see below:
"Results presented in this paper are the first direct long-term measurements of size-resolved cloud residual number concentrations of Arctic low-level clouds. It is also the first cloud residual data set that covers more than a full annual cycle, in the Arctic and globally.

We conducted a thorough evaluation of the GCVI measurements by comparing them to cloud particle size distributions as measured by an FM-120 fog monitor, as well as to total particle size distributions measured behind a whole-air inlet. We derived a correction factor for the cloud residual measurements based on the cloud particle data and the experimentally determined sampling efficiency of the GCVI. For warm clouds, we could also derive a correction factor by comparing cloud residual and whole-air accumulation mode particle concentrations (under the assumption of liquid droplet activation with no size-dependent chemical composition), and we found that both methods agreed remarkably well (within one standard deviation). Our data set includes the winter months, when Arctic warming is most pronounced and clouds are hypothesised to play a key role. However, as it turns out, the winter months are not entirely straight-forward to analyse. We identified a group of data at cold temperatures where the cloud residual and cloud particle concentrations did not agree well. It is likely that this is a result of snow or ice crystal shattering artefacts. However, these points are a small percentage (∼7–8%) of the total data and the majority of the data are not affected by potential sampling artefacts.

Our measured cloud residual number concentrations generally follow the typical annual aerosol cycle previously reported for this site. For pure liquid clouds (T> 0°C), we observed activation diameters ($D_{50\%}$) in the range of 58–78 nm. The cluster analysis of cloud residual size distributions showed a $D_{50\%}$ dependence on updraft for liquid clouds (clusters 3–5). There was also a clear correlation with the total number concentration of aerosol particles. Although we cannot clearly disentangle the influence of these parameters, our analysis indicates that it is perhaps more likely that the number concentration drives the difference in $D_{50\%}$. A clear relationship between a decreasing total particle number concentration and a decrease in $D_{50\%}$ was also observed when the cloud residual size distributions were binned by temperature.

In April–October, 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. In November–March, we found, in relative terms, a significant contribution of Aitken mode particles to the cloud residual number concentration. However, the presence of the ice phase and snow complicates matters. The mode of smallest particles we observed (cluster 1) is most likely due to artefacts of crystal shattering within the wind tunnel of the GCVI or caused by fragments of cloud nuclei or scavenged particles created through secondary ice multiplication processes. With our instrumental set up, the contribution of these different processes cannot be confidently quantified. As far as cluster 2 is concerned, while artefacts and ice processes cannot be completely ruled out, we believe that the majority of the signal is real and shows new experimental evidence of the activation of aerosol particles down to ~ 20 nm in the Arctic, confirming results from previous experimental and modelling studies.

In-situ sampling of cloud droplets and ice crystals are a complex challenge. Detailed cloud phase measurements, i.e. the ratio of ice crystals to liquid droplets within the cloud and close to the GCVI, using a more sophisticated cloud probe will be needed to better understand the relative importance of CCN / INP and the importance of other related in-cloud processes. 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. In addition, detailed and size-resolved chemical composition measurements of the sampled cloud residuals and the contribution of supermicron particles would help to better understand the sources and processes related to low-level Arctic cloud formation."

Given all the changes, we also refined our abstract (incl. toning down the aspect of nanoparticles, see comment by same reviewer 1 above). The new abstract reads: "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. 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 fall and winter months. We observed activation diameters in the range 58–78 nm, but frequently also observed cloud residuals around 20 nm or smaller. 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."

1.2 Specific Comments:

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

We agree and have added to this particular paragraph the sentence:
"Satellite data show that Ny-Ålesund is located in a region with highest cloud cover in the Arctic (Cesana et al.; Mioche et al., 2015)."

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

The CVI size-dependent transmission efficiency was determined by Shingler et al. (2012). The efficiency used in this work is from their Figure 4 (grey squares). For clarity, a figure with the used transmission efficiency from Shingler et al. (2012), the extrapolation, and the average cloud particle size distribution during our measurement period has been now added to the supplementary material (see Fig. S4 in revised SI) and is being referred to within the result section.

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.

This information has been added to the manuscript: "In the overlapping size range, the size distributions from DMPS 2a and DMPS 2b were combined by using the data from DMPS 2a in all overlapping bins except the last three. DMPS 2a data were preferred because DMPS 2a is shorter than DMPS 2b, and therefore suffers fewer losses. The last three bins, however, were not corrected for multiple charges, and therefore we used the data from DMPS 2b for those bins instead." See also Fig. 1 below.

In our particle size range, the loss calculation is not very sensitive to the chosen density (hence our previous approximation). The other reviewer also commented on the chosen density being too low, and we have therefore redone the loss calculations assuming a density of $1.5\,\mathrm{g\,cm^{-3}}$ instead. There are some small differences compared to before, but they are only noticeable at the upper end of the size spectrum where sedimentation and impaction losses become more important with this higher density.

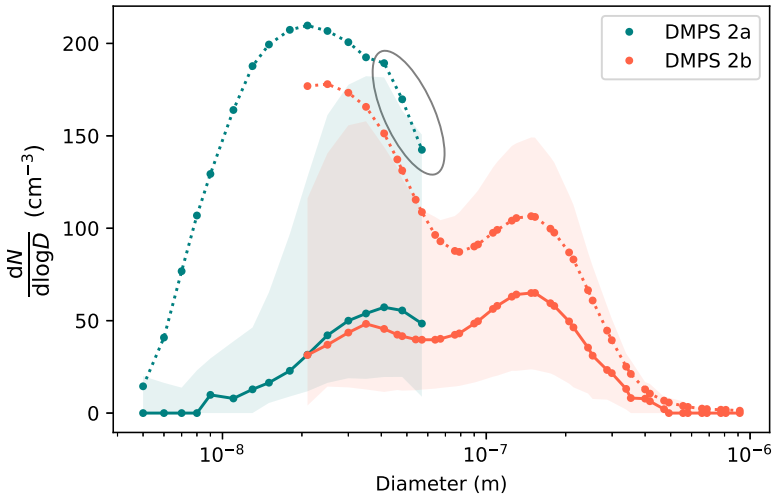

Figure 1: Average size distributions from DMPS 2a (teal) and DMPS 2b (red) for the entire data set, to illustrate how they compare in the overlapping size range. Solid and dotted lines show median and mean values, respectively, and shaded areas indicate the 25$^{\mathrm{th}}$ to 75$^{\mathrm{th}}$ percentile ranges. The bins in DMPS 2a that may sometimes be influenced by multiply charged particles (and therefore were not used) are circled.

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

Overview figures of each single day with all relevant measured parameters (GCVI data, all CPC's & size distributions, meteorology, visibility, etc.) were produced and manually checked. In addition, all lab books were screened. Suspicious periods (e.g. with spikes in the CPC's) that happened at the same time with activities outside (close to inlet) or with other maintenance work that influenced our inlet lines were removed. Other suspicious periods included periods with the GCVI detecting a cloud but with almost no particles measured by the CPC. This happened occasionally when icicles formed on the visibility sensor, which was also manifested in a smooth and almost constant visibility signal. For all these periods we also checked the centrally saved webcam images to confirm that no cloud was present. These clarifications were added to the new Sect. 2.4 (data treatment), as follows:
"The DMPS and GCVI data were processed in several steps. The logbooks from Zeppelin Observatory – which detail dates and times for visits, maintenance, instrumental issues and other observations – were examined, and data were removed when the logbooks indicated that they may be affected by the activity at the station. Next, daily overview plots of all relevant parameters were made, and each daily plot was visually inspected. Outliers (e.g. sudden concentration spikes) and suspected pollution events (e.g. concentration peaks around mealtimes or flight times) were removed. Special attention was also given to data points around gaps in the time series, and if there appeared to be issues in the data leading up to the instrument failure or after reboot, the suspicious data points were removed. Finally, several numerical filters were applied to catch additional outliers that may have been overlooked during the visual inspection. These filters looked for DMPS scans where the integrated number concentration was much higher (¿500 cm^{-3}, e.g. caused by electrical sparking inside the DMA) than the concentration measured by the total CPC, data points that showed a much higher concentration than both neighbouring data points (¿1 500 cm^{-3}; kept this high so as not to accidentally cut out nucleation events), and scans where the majority of the concentration came from the highest or lowest size bin (indicating sparks in the DMA or possible pollution).

The GCVI system outputs status codes for the operation of each part. When switching on/off of the GCVI occurred during a DMPS 1 scan, that scan was removed (since it is neither in- or out of cloud, and the enrichment factor is not defined for this case). Occasionally, there were also issues with icing of the visibility sensor, which led to the GCVI turning on despite there not being a cloud at the station. These cases were found by comparing the visibility to the measured cloud residual concentration, and data points that seemed questionable (i.e. too low concentration with respect to the visibility) were further investigated. If no cloud was detected by looking at webcam images from the station, or if the visibility was suspiciously constant (indication of icing of the sensor), the DMPS scan for those times were removed.

After the data screening, 1 729 hours of cloud residual number size distribution measurements remained. All cloud residual data are used in Fig. S3; however, for the remaining figures we were limited by the availability of concurrent data from the other instruments (DMPS 2a, DMPS 2b, the fog monitor, the ultrasonic anemometer, and the Cloudnet retrieval). Thus, slightly different subsets of the cloud residual data are used in the different figures. Table S1 shows how many hours of simultaneous measurements we have for different instrument combinations, and which figures the combinations are relevant for.

We have not applied any standard temperature and pressure normalisation or particle shape correction to the data presented here, but multiple-charge corrections have been applied to all measured size distributions. They have also been corrected for particle losses due to diffusion,

impaction and sedimentation using the *Particle Loss Calculator* by von der Weiden et al. (2009), assuming a particle density of $1.5\,\mathrm{g\,cm^{-3}}$."

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.

This may indeed be consistent with pure ice clouds. We thank the reviewer for this suggestion, and have added it as a hypothesis in the updated version of the manuscript. However, it is difficult to confirm since Cloudnet does not distinguish between cloud ice and falling snow. Due to this, as well as a comment from the other reviewer, we have now also discussed the possibility that the low particle concentrations also are consistent with shattering artefacts from snowfall (which would also explain the even lower concentrations in the fog monitor, since many snowflakes may be larger than the last channel of the FM-120). This explanation has also been added to the revised version of the manuscript (please refer to response to comment 1 above).

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.

Both reviewers raised this issue, and we agree. We have opted to remove this part from the revised version of the manuscript.

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

We have expanded the explanation to include that a supersaturation change is necessary to activate smaller particles, and this change could be caused by either an increase in updraft velocity or a decrease in particle concentrations: "The apparent $D_{50\%}$ decreases with decreasing temperature, which indicates an increase in cloud supersaturation with decreasing temperature. If the meteorological conditions are otherwise the same, this could be caused by an increase in updraft velocity or by a decrease in particle concentration (less competition for water vapour allows smaller particles to be activated). The latter is consistent with the general decrease in particle concentrations with temperature seen in the first two panels of Fig. 6. "

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

When values close to 2 are happening in the range of larger particle sizes, the concentrations are very low as can be seen in panel b. Dividing small numbers by small numbers can easily lead to values above 1. At the smallest sizes, it could be an indication of shattering. The ratio fluctuations could also 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. We have added these explanations to the revised manuscript: "The ratios are occasionally above 1 which, at the upper end of the particle size range, could be caused by small number statistics (i.e. ratios of small numbers). 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. "

Adding shades of uncertainty would unfortunately make the panel unreadable, but we have

updated the other figure that includes activation ratios (Fig. 12 in the revised version) to include the ratio of both means and medians, in order to better illustrate the range of uncertainty. We have also added an additional supplementary figure that shows the mean, median, and interquartile range of the activated fraction of particles > 100 nm as a function of temperature (similar to Fig. 7 in Verheggen et al. (2007); see Fig. 2 below).

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

The whiskers on the boxplots extend to the last data point within 1.5 times the interquartile range from the nearest quartile (the Tukey original boxplot definition). Data points outside this range can be marked as individual points, or, as done here, excluded from the plot. We have added this information to the figure captions: "The whiskers extend to the farthest points that are within 1.5 times the interquartile range from the nearest quartile. Points that fall outside the whiskers are not shown."

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

The small residuals (cluster 1) is only present in around 8 % of the data and shows on average the same values of updraft as the other clusters (see Fig. 10b and 9b in the revised manuscript). As such, they are not apparent when calculating the mean residual size distribution binned by updraft.

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.

We agree and have removed Figs. 6 and 7 from the main manuscript and just focus on Fig. 8 (now 9) to describe the shape of the cloud residual size distributions.

2 Anonymous Referee #2

Received and published: 18 June 2020

2.1 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.

We thank reviewer 2 for their detailed and helpful comments.

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 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.

We agree. Within a major restructuring of our result, discussion and conclusion section, we have toned down our language. We removed the particular sentence from the abstract and also made sure to point out that our observations cannot distinguish between INP and CCN.

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 %.

As stated in the manuscript, only 8 % of the data (cluster 1) clearly showed a different shape. We emphasise that artefacts cannot be ruled out, and have expanded the discussion (including additional figures) to try to quantify when artefacts are most likely, when they are less likely, and what bearing they have on the results. For example, we now show the comparison to the FM-120 for each cluster separately (also giving the percentage of data falling within the 10:1, 5:1 and 2:1 lines), and we have also included a supplementary figure where the cluster analysis is repeated without the most suspicious data points. Please refer to the answers to Reviewer 1, major comments 1 and 2). More details are also given in the answers below.

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.

We agree. We have added the caveats concerning the secondary ice hypothesis to the discussion of the cluster analysis. Further discussions among the coauthors, sparked by comments from both reviewers, have led us to believe that at least part of the Aitken mode cloud residuals in the winter may be the result of snowflake shattering artefacts, and this discussion has also been added in the updated version of the manuscript. As the reviewer correctly points out, residuals that are the result of scavenged particles, secondary ice processes or shattering artefacts cannot be said to be driving cloud formation since the particles were not acting as cloud nuclei. We have clarified this point in the discussion about the clusters as well. Please refer to answers to

Reviewer 1, major comments 1 and 2 where we have included text from the revised manuscript concerning these issues.

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 mixedphase clouds and would be thus needed for the Mt. Zeppelin measurements when mixed-phase clouds prevail.

We agree that it would be warranted to conduct a research project dedicated to a separate sampling of ice and liquid cloud particles at Zeppelin Observatory (which we have added to the conclusions). Continuous Ice-CVI measurements for longer periods are, however, difficult to perform. Also, one should bear in mind that most ground-based ice-CVI's suffer from the same issues as the here used GCVI system: Possible break-up of ice-crystals within the wind-tunnel or the impaction plate inside the ice-CVI (E. Weingartner, pers. comm., Nov. 2020). As discussed above, we have taken great care to evaluate the validity of our overall results and further analysis/discussion has been added to the revised manuscript. One important additional analysis and comparison of the temperature-dependence of the activation ratios of mixed-phase clouds to the observations of Verheggen et al. (2007) now also shows that the observations from Zeppelin Observatory (see Fig. 2 below and in SI) agree well with those from the high-alpine site Jungfraujoch (where the ice fraction within the cloud was directly measured and and ice-CVI was used). Although the temperature-dependent activation ration of mixed-phase clouds by Verheggen et al. (2007) was determined with a whole-air/interstitial inlet system set-up, we reach a similar result by using the GCVI/whole-air inlet set-up, which overall gives us strong confidence in the validity of our results.

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.

We thank the reviewer for their really detailed and valuable comments, which helped to improve the revised version of our manuscript. As suggested also by reviewer 1, we have shifted the focus away from the Aitken mode particles being important for Arctic cloud formation and give the entire 2-year dataset more equal weight.

The reviewer suggested that we restrict our study to only focus on pure liquid clouds, i.e. discarding all data collected during the winter months. We disagree with this, but we have taken the concerns seriously and have majorly refocused and restructured the manuscript. Based also on the comments of reviewer # 1, we now separated the presentation of our results for warm and cold clouds and performed a major restructuring of our the result, discussion and conclusion section. We chose not to discard the cold cloud data for two main reasons:

1. We believe that our new manuscript structure makes it clear to the reader which data points are likely to be affected by artefacts and which are not, and that the toning down of the conclusions relating to those data points now makes the paper well balanced, thus resolving the issues raised by the reviewer.

2. Analysing and interpreting CVI data is not a trivial task, and we believe it is of great importance and value to the scientific community to clearly communicate the types of issues that may be encountered when carrying out observations like these. Refraining from showing and discussing "suspicious" data would mean withholding potentially valuable knowledge. By being transparent and openly and honestly discussing potential problems, we hope to make life easier for others who might wish to carry out similar observations.

We openly address and quantify the issue of potential artefacts and the need for further more detailed observations dedicated to the cloud phase. As discussed in more detail above (and Figure 2 below), we now show that our observations do generally agree well with observations of mixed-phase clouds by Verheggen et al. (2007).

All in-situ cloud observations (ground-based or airborne) are unavoidably affected by instrumental and set-up-specific choices. The FM-120 is indeed influenced by wind direction and particle shape. This is true for many other airborne and ground-based cloud and aerosol sensors (Baumgardner et al., 2017). The here used FM-120 was thoroughly evaluated by Koike et al. (2019) and by own analysis and no clear bias from the wind direction was observed at Zeppelin observatory. The sampling efficiency of the GCVI determined using FM-120 data and determined from comparing the accumulation mode concentration of the cloud residual and total size distributions for warm clouds delivered almost the same factor (see revised manuscript and details below). We have added a supplementary figure (Fig. S7 in revised SI) where the cloud residual and cloud particle number concentration comparison has been separated by wind direction. In general, no clear influence of the prevailing wind direction on the concentration comparison can be observed. As such we are convinced that the FM-120 measurements are trustworthy and valid for most parts. The here used CVI system has been thoroughly evaluated in various wind-tunnel studies and in over 30,000 in-flight droplet size distributions/residue concentration intercomparisons (e.g., Shingler et al., 2012, and references therein). Given the thorough evaluation of the GCVI within previous and the here presented work, and the now provided balanced and newly structured presentation and discussion of our results, we hope that we have convinced the reviewer about the validity and novelty of our work.

2.2 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.

It is a good point, but with all the different correction factors and size cuts that occur in the different concentrations, it is difficult to device an abbreviation system that is not overly cumbersome. We hope that the restructuring and revisions of the manuscript will help make life easier for the reader.

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".

We agree and have added the missing information to the text and the revised manuscript/SI (see comments below).

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.

Computational fluid dynamics modelling of the GCVI wind tunnel flow field demonstrated that droplets smaller than roughly 60-70 micron diameter followed the flow streamlines and reached the 100 m/sec velocity within the wind tunnel throat upstream of the CVI tip. Under these findings, the sampling efficiency of the CVI inlet that was previously measured in an aerosol/droplet wind tunnel and validated by over 30,000 in-flight droplet size distribution/residue concentration intercomparisons, as outlined by Shingler et al. (2012), applies. As part of a separate in-house GCVI characterization project by the manufacturer a small cloud chamber was constructed and the GCVI wind tunnel sampled droplets from the chamber in parallel with an FSSP. Agreement between the CVI enhancement factor-corrected FSSP droplet number concentrations above the CVI cut size and the residue particle concentrations measured downstream of the CVI by a MCPC were within experimental uncertainty, typically 25% (F. Brechtel, Oct. 2020, pers. comm.). We have summarised this information in the main manuscript as follows: "Shingler et al. (2012) measured the sampling efficiency of the CVI inlet in an aerosol/droplet wind tunnel, and it has been validated by over 30 000 in-flight droplet size distributions / cloud residual concentration intercomparisons. Computational fluid dynamics modelling and a separate GCVI characterisation project by the manufacturer in a small cloud chamber showed that the Shingler et al. (2012) sampling efficiency applies to the GCVI; agreement between the corrected droplet number concentrations above the GCVI cut-size and the cloud residual particle concentrations measured downstream of the CVI by an MCPC were within experimental uncertainty, typically 25% (F. Brechtel, Oct. 2020, pers. comm.)."

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

We have harmonised the order of figures in the SI with the order in the main manuscript.

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.

We agree with this, and have taken it into consideration when restructuring the manuscript. The revised/remaining parts of these sections are now included as independent results rather than as part of the GCVI assessment.

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

The appearance or termination of a cloud event can be caused by a change in air mass with differences in aerosol properties. As such, we refrain from an additional analysis of particle size distributions before and after the cloud event (cloud residual and whole-air) since we believe that the two-year (concurrent) dataset stands for itself (over 1300 hours of observations).

We thank the reviewer for the excellent suggestion of including an alternative way to determine the GCVI sampling efficiency for liquid clouds. This has now been incorporated into the section about the GCVI assessment (which, additionally, has been divided to consider warm and cold clouds separately in line with other remarks from both reviewers). This additional analysis gave similar results as the efficiency determination using the FM-120 which further increased confidence in our observations. We include the text relating to the reviewer's suggestion here for easy access, and ask the reviewer to refer to Fig. 4 in the revised manuscript): "For warm (liquid) clouds, the sampling efficiency could also be estimated by scaling the cloud residual size distribution to the total particle size distribution under the assumption that all accumulation mode particles activate into cloud droplets. We compared cloud residual and total particle concentrations (30 min mean values) integrated above different accumulation mode threshold diameters, D_{cut}, in the range 41–505 nm. Figure 4a shows an example scatter plot for $D_{\mathrm{cut}} = 123$ nm, with the corresponding ODR fit parameters (grey data points correspond to cold clouds and are not included in the fit). Figure 4b–c shows ODR best fit slopes and coefficients of determination for all the different D_{cut} diameters (results from cold cloud data are included for completeness, but not used since we then cannot assume liquid droplet activation).

Above approximately 100 nm, the slope plateaus around 0.46. Given that this method does not take into account the cloud particle cut-size of the GCVI inlet, we can expect this estimate of the transmission efficiency to be a little lower than the one derived from the fog monitor data (0.51). A value of 0.46 is just within the standard deviation of the ratio from Fig. 3, so these two independent methods agree remarkably well. To be conservative, and to be consistent with the cold clouds (next subsection), we have corrected all (warm) cloud residual size distributions and concentrations by a factor of 2 (from Sect. 3.2.1 onward) assuming cloud residual size and cloud particle size are not correlated."

2.3 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.

We agree and have added "...and cloud processes" following the "cloud formation". In addition, we clarified on the contribution and origin of cloud residuals in the introduction and method section.

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 "groundbased counter-flow virtual impactor inlet system" and its peculiarities.

We agree and have modified this part:
"To continuously sample cloud droplets and ice crystals and to separate them from non-activated aerosol, a ground-based counter-flow virtual impactor inlet system (GCVI) was used. A detailed evaluation of the here used GCVI system is also presented."

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.

We agree and have re-written the last part of the abstract that now also addresses the changes made during the review process. Please see response to reviewer 1 above (major comment 9).

L.16: The reference should be in brackets.

Changed accordingly.

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?

We have clarified this sentence. It now reads: "Crucially, the autumn and winter seasons are also when Arctic amplification is most pronounced (Serreze and Barry, 2011; Maturilli and Kayser, 2017). This, in combination with the low background particle concentrations, makes the Arctic autumn and winter seasons more likely to experience large relative changes in aerosol particle concentrations and, consequently, changes in cloud properties."

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

Agreed and deleted. Since the size cut between Aitken and accumulation mode is not sharp, we used $\gtrapprox$ and $\lessapprox$ instead of $>$ and $<$, respectively.

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

Nucleation scavenging. We have updated the text.

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

This relates mainly to an increase in accumulated precipitation. We have added to this sentence, which is also shown by Tunved et al. (2013) in their Figures 14-15 (reference is given at the end of the sentence): "...(i.e. precipitation)..."

L.50-51: delete "for example".

We have deleted it.

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

This has been changed.

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

True. We added "and 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.

We agree and removed the word here and throughout the manuscript where appropriate.

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.

We agree and have removed the redundant parts.

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.

Parts of the remaining sample flow were used for other instrumentation from Stockholm University and collaborators which are not part of this manuscript. We added the following sentence to the instrument section: "It should be noted that other instrumentation besides the ones used in this study, and listed in Table 1, were operated during the years 2015 and 2018 behind the GCVI. That is why we used the overall high sample flow."

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.

The larger crystals and graupel that would be expected to break-up and shatter in the trap inside the CVI inlet would already have done so upstream of the CVI in the wind tunnel because they

are too large to follow the streamlines all the way to the CVI tip. Whether an impact can produce shattered crystals can be estimated using the non-dimensional Weber number (proportional to air density $\times$ velocity^2 $\times$ diameter). Sampling conditions with critical Weber number values between 10 and 12 are expected to produce fragments (Twohy et al., 2003). In the CVI these conditions occur at 100 m/s air speed and droplet diameters between 70 and 100 microns. If the shattered crystal fragments were larger than the CVI cut size they could be sampled by the CVI, however, a significant fraction of these fragments would likely not be aligned with the streamlines and would not enter the CVI. If particles would shatter inside the inlet, such events are typically clearly seen in timeline data of cloud residual particle concentration, where much higher concentrations of residual particles are observed by particle counters downstream of the CVI. Over the 11 years of deployment of over 20 CVI inlets on research aircraft and the last 7 years of deployment of the ground-based version of the CVI, artefact particle production relating to the particle trap has not been observed (F. Brechtel, Oct. 2020, pers. comm.).

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.

WMO's definition of fog can be found on the WMO's website (`https://cloudatlas.wmo.int/en/fog-compared-with-mist.html`). The current version of the "International Cloud Atlas: Manual on the Observation of Clouds and Other Meteors" from 2017 can only be found online. It can be found in various technical reports of WMO. The latest one is WMO (2008), where it e.g. states: "In climatological summaries, however, all occasions of visibility of less than 1 km are regarded as fog." We added this reference and the online version to the revised manuscript. In addition we now state the corresponding range of LWC as determined by the FM-120 by adding the following sentence: "This threshold corresponds to a large liquid water content (LWC) range of 0.0004 gm^{-3} to 0.10 gm^{-3} (5th and 95th percentile) with a median value at 0.01 gm^{-3} as measured by the FM-120 (see below) for liquid clouds (temperatures above 0°C)"

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?

The second total CPC is mainly a back-up CPC in case the first total CPC is failing. For the number concentrations shown in the manuscript we used the integrated and loss corrected particle number size distributions. This was to make sure the data were comparable, since the total CPCs of the different DMPS systems are not all the same model and therefore have different cut sizes. However, when comparing the residual number concentrations to the cloud particle concentrations, the total CPC (behind the GCVI) was used.

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.

The choice of density has no significant influence (hence our previous approximation), but we agree that 1.5 g cm^{-3} is a more realistic value and have redone the loss corrections with this

density instead. The changes are only marginal and mainly affect the larger end of the DMPS size range.

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.

Contamination means influence of particles from local activities such as maintenance on the roof of the station, the inlet system or other instruments. Our data screening involved that overview figures of each single day with all relevant measured parameters (GCVI data, all CPC's & size distributions, meteorology, visibility, etc.) were produced and manually checked. In addition, all lab books were screened. Suspicious periods (e.g. with spikes in the CPC's) that happened at the same time with activities outside (close to inlet) or with other maintenance work that influenced our inlet lines were removed. Other suspicious periods included periods with the GCVI detecting a cloud but with almost no particles measured by the CPC. This happened occasionally when icicles formed on the visibility sensor, which was also manifested in a smooth and almost constant visibility signal. For all these periods we also checked the centrally saved webcam images to confirm that no cloud was present. These clarifications were added to the new Sect. 2.4 (data treatment), see also response to Reviewer 1, specific comment L165, where we have included the text.

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.

We do apply the loss correction, perhaps the wording was unclear. What is meant is that, while the correction is applied, there are cases where it has no effect (namely when the concentration in the bin in question is zero). We recognise that this is a trivial statement that only causes confusion, and have therefore removed the sentence.

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.

Koike et al. (2019) performed thorough analysis of number concentration of cloud elements as a function of wind speed and direction (page 1802 and Appendix A). They did not observe any signature of enhanced particle losses and there is no clear difference between number of cloud elements, neither cloud water content for data observed in northerly and southerly winds. Following the reviewers suggestion, we have added a supplementary figure (Fig. S7 in revised SI) where the cloud residual and cloud particle number concentration comparison has been

separated by wind direction. In general, no clear influence of the prevailing wind direction on the concentration comparison can be observed. Together with the results from Koike et al. (2019), we believe this supports using data from all wind directions.

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.

Changed accordingly.

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

We originally did not correct it because the reference sensor is at a different altitude compared to our ultrasonic anemometer and the temperature sensor in the GCVI was broken for most of the measurement period. However, we have now analysed and compared the GCVI temperature to the uSonic temperature during the period when the former was not broken, and found that the uSonic temperature was, on average, 2.6°C higher than the GCVI temperature. We have subtracted this offset from all the uSonic temperature data presented in the revised manuscript. We added the following sentences to the revised manuscript: "In the Arctic, this temperature difference was larger. The GCVI inlet has its own temperature sensor, but it was only working for a few months at the start of our measurement period. During the overlap period, the difference between the measured acoustic temperature and the ambient temperature measured by the GCVI temperature probe was around 2.6°C. Thus, we have subtracted 2.6°C from all temperatures measured by the ultrasonic anemometer."

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.

Please see comment above (L.183) and new Fig. S7 in revised SI.

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.

We agree. A new figure (see Fig. S4 in revised SI) with the used transmission efficiency from Shingler et al. (2012), the extrapolation, and the average cloud particle size distribution during our measurement period has been added to the supplementary material and is referenced within the text.

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

We added to the method section (CVI description): "The dew point of the dry counterflow produced by the dry air generator was -40°C. "

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.

We agree and have removed this expression since it is not really needed.

L. 231: The expression "corrected cloud particle concentrations" is badly chosen and misleading. "The GCVI sampled cloud particle concentrations according to the transmission correction" or something similar would make it clearer.

We agree and have changed this sentence to: "The cloud particle concentrations, integrated above the GCVI cut-size and multiplied by the GCVI sampling efficiency, were compared ..."

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.

We added this information to the revised manuscript: "The second group [i.e. the points below the 1:10 line] is associated with very low cloud particle concentrations ($1 \pm 3\,\mathrm{cm}^{-3}$, total concentrations without cut-size and Shingler et al. (2012) correction) and the cloud particles are also fairly large in size ($11 \pm 4\,\mu$m effective radius)."

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.

Thank you for this comment. The other reviewer said "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." In their view, it seems that shattering is not necessary for more than one residual to be released. We have not been able to find any references that describe how rimed or scavenged particles would behave on an evaporating ice crystal. In the revised version of the manuscript, we have removed the Mertes et al. (2007) reference and weakened our statements to be more speculative in nature. However, if the reviewer has any references in mind that would help elucidate this issue, we would be very happy to receive them and add them to the final manuscript. The part about secondary ice particles also being seen in the fog monitor has also been clarified in the new version of the text.

The revised paragraph now reads: "Riming or impaction scavenging of interstitial aerosol particles onto an ice crystal may be able to result in more than one cloud residual emerging from the crystal as it dries inside the GCVI inlet. If more than one residual could be released through this process without the need for crystal break-up, it could be an alternative explanation that is consistent with the difference in cloud residual and cloud particle number concentrations. However, this is speculative since no strong experimental evidence exists on how rimed particles would behave inside the CVI flow regime."

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.

Thank you for spotting this. The Lauber-reference referred to the second part of the sentence about liquid droplets that can eject material when freezing as shown by Lauber et al. (2018). This sentence has been removed from the revised version.

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

Changed accordingly.

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.

We agree. In addition, one should keep in mind that the cloud particles that break within the wind tunnel of the GCVI need to produce large artefacts (above the cut-diameter of the CVI) in order to be sampled. Break-up after the stagnation plane of the CVI is less likely and has not been observed. We have looked again at periods of our data when the cloud residual size distributions were dominated by cluster 1 (which we attributed to clouds with contributions of ice and snow). The total CPC data behind the CVI did not show any suspicious spikes which could be an indication of crystal shattering within the CVI (or the following sampling line). As such, if crystal shattering occurred, it most likely happened within the wind tunnel above the CVI inlet. Also in reference to comments by reviewer 1, we have clarified these aspects and changed this paragraph (please refer to the response to Reviewer 1, Major comment 1).

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

Yes, good point. We have added this.

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.

This is true, and we have added discussion about the potential influence of snowflake shattering artefacts in the revised manuscript. See response to Reviewer 1, major comment 1. We agree with the reviewer that it is a likely explanation. That said, also bear in mind that the fragments need to be larger than the GCVI cut-size and aligned with the streamlines in order to be able to enter the inlet and be sampled. The wind tunnel also has a rain cover to avoid rain droplets entering the inlet.

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

We agree and removed the reference (although the same loss mechanisms for droplets are also relevant for ice crystals).

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.

Both reviewers raised this issue, and we agree. We have opted to remove this part from the revised version of the manuscript. A sensor that discriminates the cloud phase was unfortunately not available and it is clear that these measurements would be really valuable.

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!

We agree that our wording sounded too insecure at this point. With the new analysis and the re-structuring of this section (warm and cold clouds separately), we improved this part.

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.

A figure with the used transmission efficiency from Shingler et al. (2012), the extrapolation, and the average cloud particle size distribution during our measurement period has been added to the supplementary material (see Fig. S4 in revised SI).

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

We really appreciate the high detail the reviewer has commented which clearly helped to improve our manuscript. However, statements like "for the sake of convenience", "funny conclusion" or "dubious and a bit helpless" are neither encouraging nor constructive.

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

Yes, we agree. This has been fixed.

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.

We have expanded the explanation to include that a supersaturation change is necessary to activate smaller particles, and this change could be caused by either an increase in updraft velocity or a decrease in particle concentrations: "The apparent $D_{50\%}$ decreases with decreasing temperature, which indicates an increase in cloud supersaturation with decreasing temperature. If the meteorological conditions are otherwise the same, this could be caused by an increase in updraft velocity or by a decrease in particle concentration (less competition for water vapour allows smaller particles to be activated). The latter is consistent with the general decrease in particle concentrations with temperature seen in the first two panels of Fig. 6. "

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.

We have removed the part about the EF uncertainties as we agree that there should be none (unless you count uncertainties in flow velocities). We have added some alternative explanations, so the text now reads: "The ratios are occasionally above 1 which, at the upper end of the particle size range, could be caused by small number statistics (i.e. ratios of small numbers). 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."

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.

Here the reviewer most likely misunderstood our arguments. The whole air inlet is very different from the GCVI inlet. Also the flows, velocities and the way particles and clouds are moving inside these two inlets are very different. Thus it is very unlikely that both inlets will produce the same artefacts with a similar frequency and size distributions. We have clarified this statement and moved it to the discussion concerning the clusters (where we have consolidated most of the discussion about artefacts and alternative origins of the small residuals). The statement is not meant to be understood as "there are no artefacts", but rather as "we see the same size particles in a different inlet that should not suffer from the same types of artefacts, therefore there may be other explanations for the small particles (in addition to artefacts)."

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.

Please see the answer to the previous comment.

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.

Cluster 1 (the small residuals) is only present in around 8 % of the data and shows on average the same values of updraft as the other clusters (see Fig. 10b and 9b in the revised manuscript). As such, the "cold cases" are not apparent when calculating the mean residual size distribution binned by updraft, and therefore not apparent in the calculated ratios.

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

This has now been added.

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?

The reviewer probably has misread the text. In the original text in lines 375-376 it is written "peak concentration" and not "peak saturation". The differences related e.g. to the monthly maximum in peak concentration, which other studies have observed in other months. These differences come natural due to the natural annual variability in aerosol properties. For clarification, we have changed this sentence to: "There are some differences to previous measurements at Zeppelin Observatory related to the natural variability of aerosols, for example in terms of when peak concentration were observed."

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.

Agreed, we wanted to indicate that there is relatively good agreement between the GCVI and the fog monitor. We have reworded the sentence to make it clearer: "As seen in Fig. 2a, the shape and the magnitude of the cloud residual annual cycle agree nicely with 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.

We have removed this expression in the revised version of the manuscript.

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

We have clarified this sentence, so that it now reads "number concentration of both cloud residuals, cloud particles, and total particles".

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.

We agree that it is a pity, and hopefully future studies can shed more light on this. We have included this line of argument in the discussion of artefacts in clusters 1 and 2, with the sentence "Large crystals are expected to be more prone to shattering, and indeed clusters 1 and 2 are related to larger cloud particles than the other clusters (Fig. 9a)." See also response to Reviewer 1, major comment 1 for further changes to the text.

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)?

Both measurements are concurrent and in the text we have changed "ambient" for "total" now (which includes interstitial particles and residual particles resulting from dried cloud particles).

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.

We have expanded the discussion and include the possibility that these clusters are affected by artefacts. However, in contrast to what the reviewer suggests, we believe Cluster 2 is influenced to a much lesser extent than Cluster 1.

Further discussions among the coauthors, sparked by comments from both reviewers, have led us to believe that at least part of the Aitken mode cloud residuals in the winter may be the result of snowflake shattering artefacts (most likely in the wind tunnel of the GCVI), and this discussion has been added in the updated version of the manuscript. We suspect snow because particles need to reach a certain size for shattering to be likely, and this size ($\sim 70\,\mu$m) is far larger than the main mode seen in the FM-120. There may be a few cloud droplets or ice crystals that are large enough to shatter but, as the reviewer has pointed out in several comments, such particles should be far outnumbered by the main droplet mode. Thus, they would have to be very large indeed to shatter into enough fragments to significantly affect the measured cloud residual distributions (bearing in mind that fragments still need to exceed the GCVI cut-size and be aligned with the streamlines in order to be sampled). Cluster 1, which is linked to cold temperatures has no clear clear droplet mode, is therefore likely to be influenced by snow shattering artefacts. Cluster 2, on the other hand, has a clear droplet mode and has a temperature distribution (see Fig. 10f in revised manuscript) that is not appreciably different from those of clusters 3–5. In addition, its shape is not so much flat as bimodal with two broad modes. However, it is possible that the influence of artefacts gives the size distribution a flatter appearance — when the cluster analysis is repeated without the most suspicious data points (new supplement Fig. S12) the bimodality can be seen more clearly, which supports this hypothesis. For all clusters, we now provide separate scatterplots of number concentration of residuals versus cloud particles. In this way, the reader can easily assess how much data of the individual clusters is potentially affected by artefacts. See also response to Reviewer 1, major comment 1 (for text from the revised manuscript relating to these issues).

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.

Agreed. With the restructuring of the manuscript, we now show the annual cycle of number concentrations separately for liquid-dominated clouds and cold clouds. We have modified the text to point out that the comparison to the CCN counter studies can only be made for liquid clouds: "Since these other techniques cannot measure INP, we will only compare them to our liquid cloud-dominated data. "

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

Agreed, we have changed the text accordingly.

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.

This statement has been removed.

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).

We agree that the explanation was incomplete as far as the CCN-limited regime goes. We have completely rewritten the conclusions and this text has been removed. Schwarzenboeck et al. (2000) analysed data where the aerosol size distribution was dominated by accumulation mode aerosol or accumulation mode aerosol contributed significantly to total aerosol number concentration. For these conditions the reviewer is correct with this statement. However, in our case we observe approximately one order of magnitude lower aerosol concentrations and relatively larger contribution of Aitken mode aerosol. Unfortunately we do not have information about the chemical composition, but a plausible explanation can be also that chemistry related hygroscopic properties can play much more important role than in the case of observations from Puy de Dome reported by Schwarzenboeck et al. (2000), and in this context the argument that only few percent of Aitken mode can be activated may not hold.

L. 495-499: Again, the argumentation her is only partly true. Findings from mixedphase 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.

In the revised version of the manuscript, we discuss liquid clouds separately from mixed-phase and ice clouds, so the citations no longer appear together in the same sentence. The conditions with respect to aerosol number density, shape of size distribution and environmental conditions (ambient T close to or above 0°C) in Schwarzenboeck et al. (2000) are comparable with our spring and summer conditions. Here our data and analysis agree well with their work. They did not present any observations for cases with ambient temperatures well below zero, nor at aerosol concentrations as low as those we observed during periods when the Aitken mode dominated the cloud residual size distribution. With regard to the observations of Mertes et al. (2007) using an ice-CVI, it is true that this instrument removes all larger cloud droplet. However, it is interesting to note that our cluster 1 does not show a droplet mode and as such we suspect the cloud to consist of pure ice (and snow) and as such might be comparable to the observations of Mertes

et al. (2007). It is therefore still valid to make the link of cluster 1 to previous observations of residual size distributions resulting from ice particles.

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?

We observed Aitken mode particles, and do provide a discussion on some plausible sources (e.g. see next comment). In addition, we highlight that future work on the origin of these particles is needed (e.g. using more sophisticated aerosol mass spectrometry measurements, see next comment). Studying the origin of these small particles is not within the scope of this paper.

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.

This has been addressed in the revised version of the manuscript. See earlier answer to major general comment C, as well as response to Reviewer 1, major comments 1 and 2.

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.

This is part of the discussion and various hypotheses are presented. The shape of cluster 1 has been observed by CVI measurements within cirrus clouds (Seifert et al., 2003) and by ice-selective inlets (Mertes et al., 2007). It should be clear to the reader that this discussion of the various possible scenarios is done in the subjunctive form. As mentioned above, we have added further analysis and changed the discussion. The increased contribution of ice is also supported by the independent Cloudnet observations (see now Fig. 11). In addition, we have now compared our observations the study of Verheggen et al. (2007), which we missed in the previous version of our manuscript. Indeed, our observations of the activated fraction of mixed-phase clouds agrees surprisingly well with the observations of mixed-phase clouds at the high-alpine site Jungfraujoch (see their Fig. 7 and Fig. 2 below) giving further support to our findings. Verheggen et al. (2007) derived similar activation ratios although using observations from an interstitial and whole-air inlet, while our findings are based on a more complex sampling via the GCVI inlet. Still, it is surprising that we find a similar shape of the activated fraction with a similar maximum at 0 °C. Verheggen et al. (2007) had more sophisticated cloud probes available and could link this decreasing activation ratio with decreasing temperature to the increased contribution of ice crystals which is consistent with the Wegener-Bergeron-Findeisen process taking place inside mixed-phase clouds. This is consistent with a decreased amount of activated accumulation mode particles seen in cluster 1. For our study, it would have been favourable to perform more detailed measurements e.g. with ice-selective inlets and especially with more advanced cloud probes that are capable of distinguishing between cloud droplets and ice crystals. The findings of this work were also one motivation to deploy more sophisticated cloud probes within our current NASCENT campaign (https://www.aces.su.se/research/projects/the-ny-alesund-aerosol-cloud-experiment-nascent-2019-2020/). Here, collaborators from ETH Zürich have deployed holographic cloud probes (e.g., Henneberger et al., 2013) at Zeppelin Observatory and on a balloon to investigate the physical properties of mixed-phase clouds.

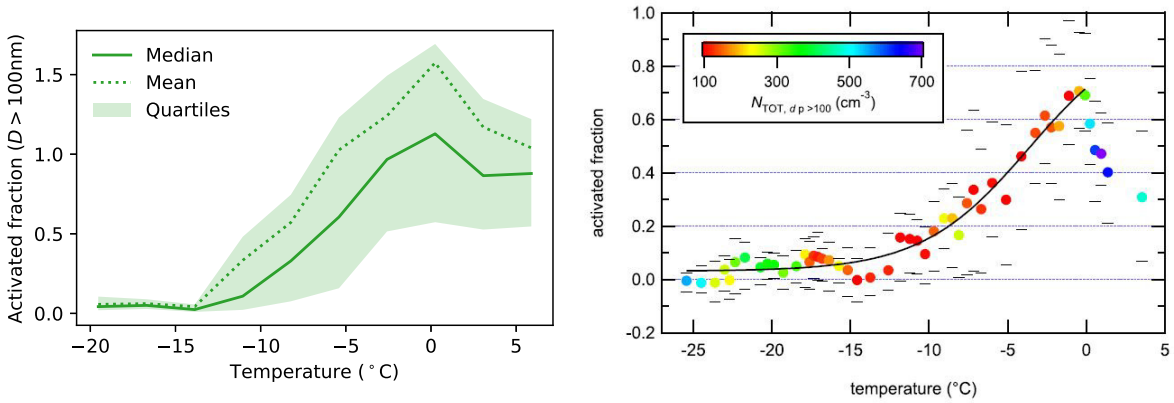

Figure 2: **Dependence of activated fraction on temperature.** The activated fraction is here defined as the ratio of cloud residual and total particle concentrations integrated above 100 nm diameter. **Left:** Our observations (ratios are based on 30 min average concentrations). Solid and dotted lines show median and mean values, respectively, and shaded areas indicate the 25th to 75th percentile ranges. **Right:** Figure 7 from Verheggen et al. (2007). Note that Verheggen et al. (2007) determined the activated fraction using observations behind a whole-air and interstitial inlet, while our observations are based on measurements behind the GCVI and whole-air inlet. The left panel has been added to the SI of the revised manuscript.

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

We have added a new section describing the data treatment in detail. See clarifications above.

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.

First of all, clear outliers due to instrument malfunctioning and local pollution were removed (see data treatment section). In accordance with this and the previous comments, the revised version now addresses the previous mentioned shortcomings. We now also perform the analysis for cold and warm clouds separately. In addition, the cluster analysis quantifies the contribution of the different shapes of the residual size distribution. Cluster 1 and 2, which might be the most suspicious ones to the reviewer, are most likely linked to ice. As recommended by the reviewer, we repeated the cluster analysis by removing the data points with largest disagreement between FM-120 and GCVI (new Fig. S12 in supplementary material), but still the main messages almost stay unchanged. Nevertheless, we still address the issue of possible artefacts more clearly in the revised version (as they can happen with almost all cloud in-situ instrumentation). Ice crystals need to reach a certain size for shattering to be likely, and we thus believe that some of the small particles we observe may be the result of snowflake shattering in the wind tunnel of the GCVI (as the reviewer also suggested as a possibility in an earlier comment). This discussion has been added to the revised manuscript. It would also explain why the concentrations measured by the FM-120 are much lower, since the snowflakes should be larger than the last channel in the fog monitor.

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.

We have rewritten the conclusions, so the phrase referred to by the reviewer no longer occurs (see response to Reviewer 1, major comment 9 for text). In the revised version of the manuscript we have addressed caveats relating to the secondary ice hypothesis more clearly, and clearly stated that the residuals from secondary ice particles do not represent cloud nuclei (see response to Reviewer 1, major comment 2).

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

We have deleted this sentence in the revised manuscript.

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.

We did not intend to say that 100 nm was the lower size limit for the accumulation mode — we simply mentioned that the use of a fixed diameter (e.g. 100 nm) to determine which aerosol particles act as CCN is a common approach used in a number of aerosol–cloud interactions studies, and we wanted to highlight that this approach is not suitable in the Arctic.

In the revised version of the manuscript, we do not introduce the 100 nm discussion, since the figure that was related to this has been removed in favour of figures relating to the cluster analysis, based on the recommendations from Reviewer 1.

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.

Given the previous comments by both reviewers, we have removed this part and rewritten the conclusions (see response to Reviewer 1, major comment 9 for text).

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.

We agree and as such have removed this part. As summarised above, we have completely rewritten the conclusions (see response to Reviewer 1, major comment 9 for text).

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.

This was a mistake in the caption, thanks for pointing it out! It has now been fixed.

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

The original caption already included the line "The transmission efficiency of the GCVI inlet (Shingler et al., 2012) has been included in the calculation of the cloud particle number concentration in all panels." We have replaced the phrase "cloud particle number concentrations as measured by the FM-120 fog monitor" with "cloud particle number concentrations derived from the FM-120 fog monitor measurements" to make it more clear.

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.

"( ]" is standard interval notation indicating whether the endpoint is excluded (soft bracket) or included (hard bracket) in the interval. In addition, we added to the caption of Fig. 3 and 4 (see also comment by reviewer 1): "The whiskers extend to the farthest points that are within 1.5 times the interquartile range from the nearest quartile. Points that fall outside the whiskers are not shown." The mean is not shown since this standard box plot shows the median and 25th and 75th percentiles (quartiles), which should be obvious from the revised figure caption.

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

This comment refers to a figure panel which has been removed in the revised version of the manuscript.

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.

This has been fixed.

3 Further changes

- We now use the total CPC (behind GCVI) for the comparison with the FM-120 (before the integrated number concentration from the DMPS scan was used). The results have improved slightly.
- We moved Figure S11 from the SI to the main text (now Figure S10). We have added the corresponding average cloud particle concentration to panel d.
- As suggested by reviewer 1, we also moved Figure S7 from the SI to the main part (now Fig. 12). To be consistent with the other size distribution plots, we show the distributions on a log-y-scale. We also added median and percentile values.

- In addition, we have removed Figures 6, S8 and S9. These figures were replaced with the new Figure S8, which shows the monthly averages of cloud residual size distributions for all, warm and cold clouds separately.
- We removed the box plots (Figure 7 and S10) showing the contribution of small particles (box plots).

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~~The role~~ A long-term study of ~~nanoparticles in Arctic~~ cloud ~~formation~~residuals from low-level Arctic clouds

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Abstract. 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 ambient aerosol particles~~ and cloud processes, in Arctic low-level clouds measured at Zeppelin Observatory, Svalbard. ~~A detailed evaluation of the~~ To continuously sample cloud droplets and ice crystals and separate them from non-activated aerosol, a ground-based counter-flow virtual impactor inlet system ~~is also presented. Cloud residuals as small as 15 nm are routinely observed especially during the dark period and are potentially linked to ice, supporting prior work suggesting that classical droplet activation is not the only relevant process in the formation of Arctic low-level~~ (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. The ~~reported measurements and findings provide a new basis for improving our understanding of Arctic clouds and for developing robust parameterisations of mixed-phase clouds in Earth system models.~~ 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 fall and winter months. We observed activation diameters in the range 58–78 nm, but frequently also observed cloud residuals around 20 nm or smaller. 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.

1 Introduction

Aerosols and clouds are important for climate, yet they remain one of the largest sources of uncertainty in climate projections (Boucher et al., 2013). Many of the parameters that govern cloud and aerosol formation are subject to change as the climate changes as well, which further obscures the picture. The Arctic is a region of particular interest, because it is warming more rapidly than the rest of the globe (Serreze and Francis, 2006; Serreze and Barry, 2011). In terms of aerosol particles, the Arctic is characterised by a distinct seasonal cycle with low natural background number concentrations for parts of the year

Willis et al. (2018)(Willis et al., 2018). The low background concentration is especially true for late autumn and early winter when the absence of sunlight and direct particle sources inhibits natural emissions and the formation of new particles (Tunved et al., 2013). This means that small changes in Arctic aerosol particle concentrations, for example following sea ice loss and increased natural marine emissions (Struthers et al., 2011) or altered transport and/or emissions of anthropogenic particles (Law and Stohl, 2007), can potentially cause large changes in cloud properties (Mauritsen et al., 2011). Crucially, the autumn and winter seasons are also when Arctic amplification is most pronounced (Serreze and Barry, 2011; Maturilli and Kayser, 2017), which makes such changes . This, in combination with the low background particle concentrations, makes the Arctic autumn and winter seasons more likely to happenexperience large relative changes in aerosol particle concentrations and, consequently, changes in cloud properties. Due to the sparsity of observations, we know less about cloud and aerosol processes in the Arctic than elsewhere. New long-term observations are thus essential for closing existing knowledge gaps.

Long-term observations of Arctic aerosol particles generally come from a relatively small number of permanent measurement stations. While there are differences in aerosol properties between the sites, it has been shown that they all share common features both in terms of particle number concentration and particle number size distribution (Freud et al., 2017). This characteristic seasonal cycle of Arctic aerosol properties has been demonstrated previously for individual sites (Ström et al., 2003; Tunved et al., 201 During the transition from winter to springtime, the number concentration of accumulation mode particles (typically diameter > diameter $\gtrsim$ 60 nm) increases due to long-range transport of polluted air masses – a phenomenon known as Arctic haze (Mitchell, 1956). In summer, changes in circulation and cloud cover lead to efficient nucleation scavenging of these particles, subsequently lowering their concentration (Tunved et al., 2013). Lower accumulation mode particle concentrations, together with increased biological activity and photochemistry, helps facilitate new particle formation leading to number size distributions dominated by the smaller, Aitken mode particles (typically diameter < diameter $\lesssim$ 60 nm) in the Arctic summertime (Ström et al., 2003). During autumn, the particle sinks are stronger than the sources because neither transport nor new particle formation is efficient, which leads to low in the Arctic is less common and aerosol removal processes (i.e. precipitation) are stronger compared to aerosol sources, resulting in gradually decreasing aerosol number concentrations across the particle size spectrum (Tunved et al., 2013).

Studies characterising Arctic cloud condensation nuclei (CCN) generally cover short time periods, and only a couple of studies exist that look at the seasonal cycle in the Arctic (Jung et al., 2018; Dall'Osto et al., 2017; Schmale et al., 2018). Jung et al. (2018) measured CCN on Svalbard and found that the seasonal variation in CCN concentrations correlated well with the variation in accumulation mode aerosol particle concentrations. They also identified new particle formation and subsequent particle growth as contributors to summertime CCN concentrations, in line with results from a previous long-term study (Dall'Osto et al., 2017) as well as shorter airborne and ground-based measurement campaigns (Leaitch et al., 2016; Zábori et al., 2015). CCN number concentrations in the Arctic have been found to range between a few tens and a couple of hundred particles cm^{-3} (Jung et al., 2018), although concentrations vary spatially. Local concentrations of less than 1 and more than 1000 cm^{-3} have been reported (Mauritsen et al., 2011; Moore et al., 2011). CCN are of course only part of the picture – in cold and mixed-phase clouds, ice nucleating particles (INP) are also important. INP are much rarer, with concentrations several orders of magnitude

lower than typical CCN concentrations. In the Arctic, INP concentrations have been found to range between approximately 10^{-5} and 10^{-1} L^{-1} ~~(see e.g., Wex et al., 2019; Tobo et al., 2019; Irish et al., 2019)~~(e.g., Wex et al., 2019; Tobo et al., 2019; Irish et al., 20

An important caveat is that all of the aforementioned studies measure CCN and INP concentrations by artificially activating aerosol particles. It is, however, possible to study CCN and INP properties directly inside clouds, by measuring the so-called cloud residuals that remain when cloud droplets and ice crystals (collectively termed cloud particles) are dried. This can ~~for example~~ be achieved with a counterflow virtual impactor (CVI) inlet (Ogren et al., 1985; Noone et al., 1988), which separates cloud particles from unactivated aerosol particles on an inertial basis. Because the CVI can measure both water and ice particles, cloud residuals may correspond to either CCN~~or INP~~ , INP or results from in-cloud processes (e.g. secondary ice; Field et al., 2016). In the Arctic, CVI inlets have previously only been deployed during short, dedicated aircraft campaigns (McFarquhar et al., 2011; Wendisch et al., 2019) and, until now, no long-term observations of cloud residual properties have been ~~performed~~ carried out either in the Arctic or globally. Here, we present a unique ~~dataset~~ 2-year data set of size-resolved cloud residual ~~number concentrations recorded between 2015 and 2018 at Zeppelin Observatory~~ and total particle measurements recorded on Svalbard using a ground-based CVI inlet. These are the first continuous measurements of cloud residuals in the Arctic that cover the full annual cycle. Our observations are accompanied by measurements of total aerosol particles (interstitial and activated aerosol particles) and ~~ambient~~ cloud particle size distributions, meteorological parameters as well as remote sensing data, which, taken together, provide valuable new information about the elusive Arctic ~~CCN~~cloud nuclei.

2 Methods

We present total particle and cloud residual size distributions and ~~integrated~~ number concentrations measured ~~during more than 2 years (~~from 26 November 2015 to 4 February 2018 ~~)~~ at Zeppelin Observatory using two different inlet systems. These measurements are complemented by measurements of ~~ambient~~ cloud particle size distributions, temperature, wind parameters and remote sensing data which are described below. A schematic illustration of the experimental set-up and a photo of the inlet systems at Zeppelin Observatory are shown in Fig. 1. Tables 1 and S1 (in the supplementary material) give further details on the instrumentation and data coverage.

2.1 Site description

Zeppelin Observatory (78°54'N 11°53'E) is located on Svalbard in the high Arctic, approximately 2 km south of the research village Ny-Ålesund. Situated 480 m above sea level (inlet height) on the ridge of Mt. Zeppelin, the station is largely unaffected by local pollution sources and often in cloud (∼16 % of the time in 2015–2018, where *in cloud* is defined in this work as visibility < 1 km for at least 5 min as measured by the visibility sensor, see below), making it well-suited for the study of Arctic aerosol particles and clouds. Note that the observed cloud occurrence may not exactly equal the annual mean cloud occurrence at the station, as we have a slightly uneven data coverage for the different months (cf. right panel in Fig. 8 as shown later in Sect. 3.0.2).

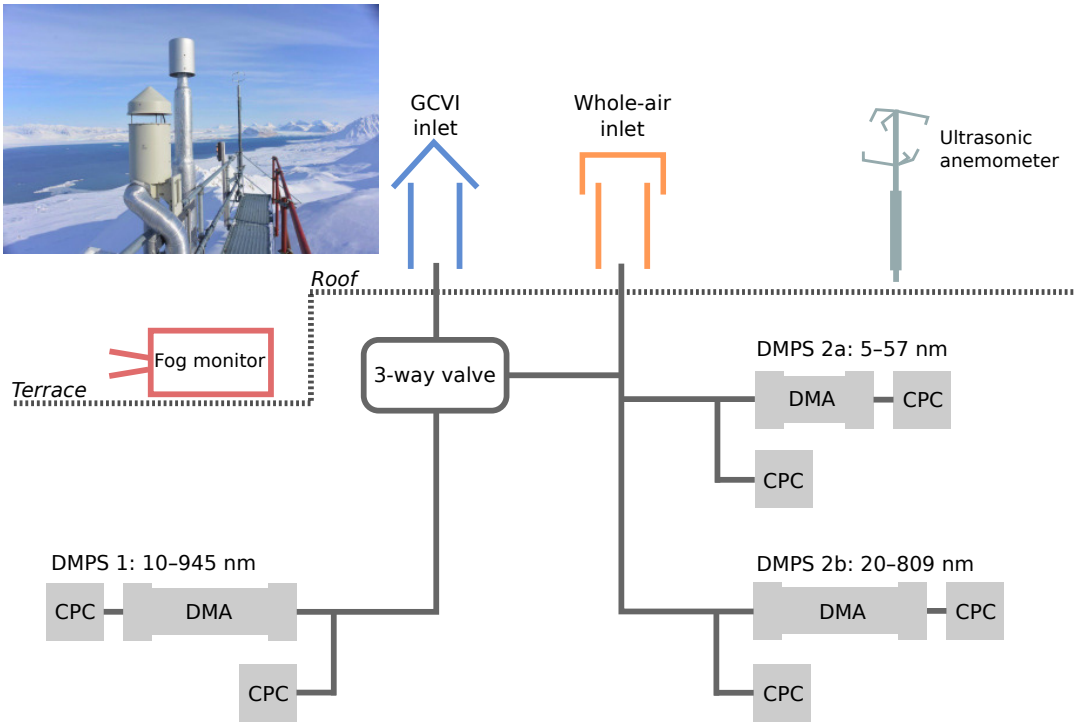

Figure 1. Schematic illustration of the experimental set-up at Zeppelin Observatory. The diagram shows how the whole-air inlet (orange) and the ground-based counterflow virtual impactor (GCVI) inlet (blue) are connected to the differential mobility analysers (DMAs) and condensation particle counters (CPCs). The 3-way valve switches the sample flow to the instruments on the left-hand side from the GCVI inlet to the whole-air inlet when there is no cloud to be sampled. Cloud sampling is activated if the visibility drops below 1 km (measured by a visibility sensor (not pictured) next to the GCVI inlet). Auxiliary measurements from a fog monitor and an ultrasonic anemometer have also been included in the data analysis.

Two predominant wind directions are characteristic for the site: south and north-north-west with a median horizontal and vertical wind speed of 3.0 ms^{-1} and 0.7 ms^{-1} during periods of cloud occurrence (see Fig. S1 in supplementary material). The annual cycle of aerosol size distribution parameters is quite predictable for the site (Tunved et al., 2013), however, the site can not be regarded as being representative for the entire Arctic. Freud et al. (2017) ~~has~~ have shown that although certain similarities in aerosol size and concentration exist between the different permanent measurement sites in the Arctic, e.g. caused by similarities in transport patterns, particle formation or removal mechanisms, distinct differences were attributed to the proximity of aerosol sources, local meteorological effects or the influence of open ocean, land areas and sea ice.

In terms of cloud cover and cloud type, it is difficult to say how representative the measurements at Ny-Ålesund and Zeppelin Observatory are for the broader Arctic. Shupe et al. (2011) have analyzed the occurrence and macro-physical properties of Arctic clouds at six observatories, including Ny-Ålesund, and found, for example, that clouds are more persistent at the far western Arctic sites. More detailed analyses of cloud radar observations from Ny-Ålesund (Nomokonova et al., 2019b; Ebell et al.,

2020; Nomokonova et al., 2019a; Gierens et al., 2020) partly confirmed results of previous studies, e.g. high ~~a~~ cloud occurrence at Ny-Ålesund in summer and autumn, but also revealed differences. For example, Nomokonova et al. (2019b) revealed a higher annual cloud occurrence at Ny-Ålesund (∼81%) than Shupe et al. (2011, ∼61%). Differences in the observed cloud statistics are likely also due to different observing instruments and methods as well as different time periods analysed. Satellite data show that Ny-Ålesund is located in a region with highest cloud cover in the Arctic (Cesana et al.; Mioche et al., 2015). The previously reported cloud occurrences are much higher than what we observe, because we are observing at a fixed altitude and thus only measure low-level clouds. The representativeness of our observational conditions for the broader Arctic cloud cover (cf. Liu et al., 2012) is unclear.

Dahlke and Maturilli (2017) showed that the synoptic flow towards Ny-Ålesund represents typical Arctic climate during the summer months, while during the winter periods large scale advection from lower latitudes is dominating in recent decades, resulting in a more maritime climate. This transition will most likely affect also cloud properties with Ny-Ålesund probably becoming less representative of the sea-ice dominated Arctic.

2.2 Inlet systems

2.2.1 Whole-air inlet

The standard aerosol inlet is heated and fulfils the World Meteorological Organization (WMO)/Global Atmosphere Watch programme guidelines for aerosol sampling of whole-air (Kazadzis, 2016) and has similar characteristics as the inlet described by Weingartner et al. (1999) which can sample cloud droplets up to 40 μm at wind speeds up to 20 ms^{-1}. It is placed on the roof of the station, and particle-laden air is brought into the lab where an isokinetic flow splitter directs the air to the different sampling instruments through quarter inch stainless steel tubing. The air is not actively dried, but the temperature difference between the outside and the inside of the lab causes a reduction in the relative humidity. During our sampling period, the relative humidity of the sample flow was always below 40 % (mean±std for our period: 13±7 %).

2.2.2 Ground-based counterflow virtual impactor inlet

For sampling of cloud residuals, we utilise a ground-based counterflow virtual impactor (GCVI; Brechtel Manufacturing Inc., USA, Model 1205) inlet, which is based on the working principles described in Noone et al. (1988). The inlet uses opposing air flows to ~~filter~~ separate out particles with low inertia (i.e. interstitial particles), so that only ~~activated~~ cloud particles (i.e. droplets and ice crystals) are sampled. A detailed technical description of the GCVI can be found in Shingler et al. (2012). Here, we outline the basic principles only.

The GCVI inlet at Zeppelin Observatory is mounted vertically on the north side of the station roof. During operation, cloudy air is accelerated onto the tip of the inlet with the help of a wind tunnel with typical airspeeds of around 120 $\mathrm{m\,s}^{-1}$ (monitored with a pitot tube). When the ambient air meets the dry counterflow within the GCVI, two stagnation planes are generated where only particles with sufficient inertia (i.e. cloud droplets or ice crystals) can pass through and enter the sample flow. The sample flow rate is set to 15 $\mathrm{L\,min}^{-1}$ by automatic mass flow controllers that take into account the actual sample flow of each

connected instrument. It should be noted that other instrumentation besides the ones used in this study, and listed in Table 1, were operated during the years 2015 and 2018 behind the GCVI. That is why we used the overall high sample flow. The lower cut-size ($D_{50\%}$) in the inlet is calculated by the instrument software and is determined by the different flow velocities and the distance between the stagnation planes. Shingler et al. (2012) compared experimentally determined cut-sizes to those predicted by the software and found good agreement. The cut-size was generally between 6 and 7 µm aerodynamic diameter during our sampling period. As the cloud particles travel through the inlet, they are dried until only the cloud residuals remain. The dew point of the dry counterflow produced by the dry air generator was -40°C. Cloud particles larger than approximately 40 µm in diameter are impacted in a particle trap inside the inlet due to their long evaporation times (Shingler et al., 2012).

The GCVI is only operated when there is a cloud at the station. The system is automated and uses a visibility sensor to determine whether or not a cloud is present. The GCVI is turned on when the visibility drops below 1 km~~(~~, which is the WMO's definition of fog (see WMO, 2008, and at https://cloudatlas.wmo.int/). This threshold corresponds to a large liquid water content (LWC) range of 0.0004 gm^{-3} to 0.10 gm^{-3} (5th and 95th percentile) with a median value at 0.01 gm^{-3} as measured by the FM-120 (see below) for liquid clouds (temperatures above 0°C). Visibility is the only criterion used, so there is no discrimination between precipitating and non-precipitating clouds. When the visibility is above 1 km, instruments that normally sample behind the GCVI inlet instead receive their sample flow from the whole-air inlet. This is achieved with a three-way valve (installed in April 2017) between the two inlets and the instruments, and allows us to collect duplicate measurements of particle size and concentrations for quality assurance during non-cloud periods.

Particles that enter the wind tunnel are concentrated ~~at~~ inside the tip of the CVI inlet, meaning the sampled air is effectively enriched in cloud particles relative to the ambient air. The concentrations observed behind the GCVI therefore have to be corrected by an enrichment factor (EF), which depends on the airspeed in the wind tunnel, the sample flow rate and the geometry of the inlet itself (Shingler et al., 2012). With our set-up, the EF was 11.9 ± 1 (median 12). It should be emphasised that even after correcting for the EF, the cloud residual concentrations measured behind the GCVI cannot be considered as absolute due to the transmission efficiency of the inlet. Because the transmission efficiency depends on the size of the cloud particles before they are dried, it cannot be corrected for. However, an estimate of the absolute cloud residual concentrations can be obtained by back-calculating from the ~~ambient~~ cloud particle size distribution (as measured by a fog monitor) using the experimentally determined cloud particle size dependent transmission efficiency (Shingler et al., 2012) of the GCVI inlet. Shingler et al. (2012) measured the sampling efficiency of the CVI inlet in an aerosol/droplet wind tunnel, and it has been validated by over 30 000 in-flight droplet size distributions / cloud residual concentration intercomparisons. Computational fluid dynamics modelling and a separate GCVI characterisation project by the manufacturer in a small cloud chamber showed that the Shingler et al. (2012) sampling efficiency applies to the GCVI; agreement between the corrected droplet number concentrations above the GCVI cut-size and the cloud residual particle concentrations measured downstream of the GCVI by an MCPC were within experimental uncertainty, typically 25% (F. Brechtel, Oct. 2020, pers. comm.).

As will be shown below, we find that on average half of the ~~ambient~~ cloud particles make it into the GCVI sample flow, and cloud residual concentrations therefore have to be multiplied by a factor of 2 to match the observations from the fog monitor (see Sect. 3.1).

2.3 Instrumentation

See Fig. 1 for a schematic overview of the experimental set-up and Table 1 for a summary of the instruments used, parameters measured and their temporal and/or spatial resolutions.

Table 1. List of instruments, measured parameters and their temporal and/or spatial ~~resoultion~~resolution.

Instrument	Parameters	Resolution
DMPS 1	Aerosol particle number size distributions (diameters 10–945 nm)	5–7 min
DMPS 2a–b	Aerosol particle number size distributions (diameters 5–809 nm)	15–16 min
GCVI	Visibility (proxy for cloud presence)	1 s
FM-120	Cloud particle number size distributions (diameters 3–47 μm)	10 s
Ultrasonic anemometer (METEK uSonic-3)	3D wind field, virtual temperature	1 s
Cloudnet	Target classification (in terms of occurrence of e.g. liquid droplets, ice crystals, drizzle, etc)	30 s, 20 m altitude bins

2.3.1 Differential mobility particle sizer

Particle number size distributions were measured with a differential mobility particle sizer (DMPS). The experimental set-up at Zeppelin Observatory has three DMPS instruments: one is behind the GCVI inlet (DMPS 1) and the other two are behind the whole-air inlet (DMPS 2a–b). DMPS 1 (sample flow 1 L min^{-1}, sheath-air flow 4.8 L min^{-1}) consists of a medium Vienna type differential mobility analyser (DMA; length 0.28 m, outer radius 0.033 m, inner radius 0.025 m) and a condensation particle counter (CPC; TSI Inc., USA, Model 3772). Another CPC (TSI Inc., USA, Model 3772) is used in parallel with the DMPS to measure the total particle number concentration. DMPS 1 is set to measure particles from 10 to approximately 945 nm in mobility diameter. A full DMPS 1 scan (small to large or large to small diameters) takes approximately 6 min to complete.

DMPS 2a and DMPS 2b measure different but overlapping size ranges. They are synchronised as one system (DMPS 2a–b) that runs on the same software. DMPS 2a (sample flow 1 L min^{-1}, sheath-air flow 9.9 L min^{-1}) measures at the smaller end of the particle size spectrum and has an extra small Vienna type DMA (length 0.053 m, outer radius 0.033 m, inner radius 0.025 m) to minimise diffusional losses, with a CPC (TSI Inc., USA, Model 3010) behind the DMA and a CPC (TSI Inc., USA, Model 3776) for measuring the total aerosol particle concentration. DMPS 2b (sample flow 1 L min^{-1}, sheath-air flow 5.2 L min^{-1}) measures the larger size particles and has a medium Vienna type DMA (length 0.28 m, outer radius 0.033 m, inner radius 0.025 m) with a CPC (TSI Inc., USA, Model 3772) behind the DMA and a CPC (TSI Inc., USA, Model 3010) for measuring the total aerosol particle concentration. Together, DMPS 2a–b span roughly the same size range as the DMPS 1, but the time resolution is approximately 15 min per full scan.

~~We have not applied any standard temperature and pressure normalisation or particle shape correction to the data presented here, but multiple-charge corrections have been applied to all measured size distributions . They have also been corrected for~~

particle losses due to diffusion and impaction using the *Particle Loss Calculator* by von der Weiden et al. (2009), assuming a particle density of 1 g In the overlapping size range, the size distributions from DMPS cm^{-3}.

After a manual data screening to remove outliers and contamination, 12a and DMPS 729 hours of cloud residual number size distribution measurements remained. The full cloud residual data are shown in Figs.2b were combined by using the data from DMPS 9 and 10; however, for the remaining figures we were limited by the availability of concurrent data from the other instruments (2a in all overlapping bins except the last three. DMPS 2a , data were preferred because DMPS 2a is shorter than DMPS 2b, the fog monitor, the ultrasonic anemometer, and the Cloudnet retrieval). Thus, slightly different subsets of the cloud residual data are used in the different figures. Tableand therefore suffers fewer losses. The last three bins, however, were not corrected for multiple charges, and therefore we used the data from DMPS S1 shows how many hours of simultaneous measurements we have for different instrument combinations, and which figures the combinations are relevant for 2b for those bins instead.

Figure S2 shows how the DMPS systems compare during non-cloud periods. The comparison is based on data collected from May 2017–February 2018 (after the installation of the three-way inlet valve, see above). In general, the instruments compare well for large particle sizes while DMPS 2a–b shows consistently higher concentrations of small particles below around 30 nm in diameter. This is to be expected, since the diffusion losses are higher for DMPS 1 due to the instrument dimensions and longer sampling lines. These diffusion losses cannot be corrected for when the concentrations in the smaller size bins are zero. Most of the differences originate from the lowest size bins between 10 and 15 nm, as can be seen in the scatter plots of Fig. S2 (panel c and d), where the integrated number concentrations of both DMPS 1 and DMPS 2a–b are shown. The slope of the orthogonal linear regression and the R^2-value (coefficient of determination) improve from 1.36 to 1.01 and 0.96 to 0.99, respectively, if particle number size distributions are integrated above 15 nm instead of 10 nm particle diameter.

2.3.2 Fog monitor

A fog monitor (Droplet Measurement Technologies Inc., USA, Model FM-120) was used to determine the ambient cloud particle size and number concentration. It uses an optical method to size individual cloud particles at a flow rate of approximately 1000 L min^{-1} (airspeed 12 ms^{-1}). The instrument is positioned facing south and measures ambient cloud particle size distributions in the size range 3.5–46 µm optical diameter (bin midpoints). More details on the instrument at Zeppelin Observatory can be found in Koike et al. (2019). It should be noted that no loss correction has been applied to the fog monitor data because no clear signatures of particle loss were found in previous work by Koike et al. (2019), although significant sampling losses were suggested in other studies depending on, for example, the cloud particle diameter, and the wind speed and wind direction relative to the fog monitor (Spiegel et al., 2012).

2.3.3 Ultrasonic anemometer

A uSonic-3 Omni (METEK GmbH) ultrasonic anemometer was used to monitor wind conditions at Zeppelin Observatory. The anemometer has 3 pairs of ultrasonic transducers arranged to form 3 paths along which the speed of sound is measured. From the difference in the travel time of sound along the 3 measuring paths, the 3D wind vector as well as the acoustic

temperature can be derived. The acoustic temperature is a close approximation of the virtual temperature, which depends on the ambient relative humidity and is generally 1–2 degrees higher than the true temperature ~~(METEK GmbH, 2013). However, note that at Zeppelin Observatory during~~ (uSonic-3 Omni Ultrasonic anemometer user manual, Metek GmbH). In the Arctic, this temperature difference was larger. The GCVI inlet has its own temperature sensor, but it was only working for a few months at the start of our measurement period~~, the median~~. During the overlap period, the difference between the measured acoustic temperature and the ambient temperature measured by ~~a Vaisala temperature probe (located in the meteorology mast at 15 m above the measurement platform) was around 3.4~~the GCVI temperature probe was around 2.6°C. ~~Nevertheless, we chose to use the temperature from our anemometer because we needed the higher time resolution that it provides~~Thus, we have subtracted 2.6°C from all temperatures measured by the ultrasonic anemometer.

2.3.4 Cloud remote sensing

The Cloudnet algorithm suite (Illingworth et al., 2007) has been applied to the Ny-Ålesund ground-based remote sensing observations from the French-German research station AWIPEV (Nomokonova et al., 2019b), which is located approximately 2 km north of Zeppelin Observatory. A standard product is the target classification which combines measurements from cloud radar, ceilometer and microwave radiometer with output from a numerical weather prediction model. Each radar height bin is classified in terms of the occurrence of e.g. liquid droplets, ice particles, rain/drizzle, melting ice and a combination of those. More details on the product for Ny-Ålesund can be found in Nomokonova et al. (2019b). For comparison with the cloud residual data collected at Zeppelin Observatory, we selected Cloudnet height bins between 400 and 600 m. We only compared cases when the cloud base height at AWIPEV was between 300 m and 600 m to ensure that the classifications were likely to be applicable also to the cloud at Zeppelin Observatory. It should be noted that this cloud base height criterion reduces the number of data points we can use, such that we only have Cloudnet data for approximately 30% of our in-cloud size distribution data.

2.4 GCVI and DMPS data treatment

The DMPS and GCVI data were processed in several steps. The logbooks from Zeppelin Observatory – which detail dates and times for visits, maintenance, instrumental issues and other observations – were examined, and data were removed when the logbooks indicated that they may be affected by the activity at the station. Next, daily overview plots of all relevant parameters were made, and each daily plot was visually inspected. Outliers (e.g. sudden concentration spikes) and suspected pollution events (e.g. concentration peaks around mealtimes or flight times) were removed. Special attention was also given to data points around gaps in the time series, and if there appeared to be issues in the data leading up to the instrument failure or after reboot, the suspicious data points were removed. Finally, several numerical filters were applied to catch additional outliers that may have been overlooked during the visual inspection. These filters looked for DMPS scans where the integrated number concentration was much higher (>500 cm^{-3}, e.g. caused by electrical sparking inside the DMA) than the concentration measured by the total CPC, data points that showed a much higher concentration than both neighbouring data points (>1 500 cm^{-3}; kept this high so as not to accidentally cut out nucleation events), and scans where the majority of the concentration came from the highest or lowest size bin (indicating sparks in the DMA or possible pollution).

The GCVI system outputs status codes for the operation of each part. When switching on/off of the GCVI occurred during a DMPS 1 scan, that scan was removed (since it is neither in- or out of cloud, and the enrichment factor is not defined for this case). Occasionally, there were also issues with icing of the visibility sensor, which led to the GCVI turning on despite there not being a cloud at the station. These cases were found by comparing the visibility to the measured cloud residual concentration, and data points that seemed questionable (i.e. too low concentration with respect to the visibility) were further investigated. If no cloud was detected by looking at webcam images from the station, or if the visibility was suspiciously constant (indication of icing of the sensor), the DMPS scan for those times were removed.

After the data screening, 1 729 hours of cloud residual number size distribution measurements remained. All cloud residual data are used in Fig. S3; however, for the remaining figures we were limited by the availability of concurrent data from the other instruments (DMPS 2a, DMPS 2b, the fog monitor, the ultrasonic anemometer, and the Cloudnet retrieval). Thus, slightly different subsets of the cloud residual data are used in the different figures. Table S1 shows how many hours of simultaneous measurements we have for different instrument combinations, and which figures the combinations are relevant for.

We have not applied any standard temperature and pressure normalisation or particle shape correction to the data presented here, but multiple-charge corrections have been applied to all measured size distributions. They have also been corrected for particle losses due to diffusion, impaction and sedimentation using the *Particle Loss Calculator* by von der Weiden et al. (2009), assuming a particle density of 1.5 g cm^{-3}.

2.5 Cluster analysis

A cluster analysis was performed to identify cloud residual size distributions that were dominated by Aitken mode particles. We used k-means clustering, implemented in the *scikit-learn* (v. 0.20.2) Python package (Pedregosa et al., 2011), which is a method to categorise data into a pre-defined number of clusters, k, where members of a cluster are as similar to each other as possible while at the same time being as different to members of other clusters as possible. Each data point is assigned to the cluster with the nearest mean. We categorised cloud residual number size distributions based on their shape, so the size distributions were normalised by the integral before applying the k-means algorithm. We selected 5 clusters ($k = 5$) to separate out the cloud residual size distributions that were dominated by the very smallest particles. Choosing fewer clusters did not fully separate this distribution of interest, while more clusters led to a further splitting of the accumulation mode (see Fig. S3).

3 ~~Assessment of the GCVI sampling efficiency~~Results

3.1 Determining the GCVI sampling efficiency

The ~~ambient cloud particle size distributions were used to evaluate the sampling performance of our GCVI system~~cloud residual concentration measured downstream of the GCVI inlet cannot be considered absolute due to the transmission efficiency of the inlet. Thus, we need to know the cloud particle number size distribution to be able to derive a correction factor for the cloud residual concentration. Assuming that the cloud particle distribution measured by the FM-120 fog monitor is an accurate

representation of the cloud particles that enter the GCVI inlet, we applied the experimentally determined size-dependent transmission efficiency from Shingler et al. (2012) (linearly extrapolated to cover the full FM-120 cloud particle size range, see Fig. S4) to calculate the cloud particle concentration above the GCVI cut-size that would have made it into the sample flow. Here, it is important to note that the transmission efficiency was determined for hollow glass beads without using the inlet counterflow (Shingler et al., 2012). As such, it does not take into account potential evaporation of water from the cloud particles in the different inlet segments. Within this work, we have only used the transmission efficiency determined for the first inlet segment, because we believe that the dry counterflow initiates evaporation which would make the transmission efficiency determined for subsequent sections an underestimation of the true transmission efficiency. This choice may result in an overestimation of the transmission efficiency (particularly of larger cloud droplets) since some losses are effectively ignored~~, but no correction is preferable to an invalid correction~~.

The ~~corrected~~ cloud particle concentrations, multiplied by the GCVI sampling efficiency and integrated above the GCVI cut-size, were compared to the ~~integrated~~ cloud residual number concentrations measured behind the GCVI inlet (by the CPC), and the result can be seen in Fig. 2. Given the uncertainties involved, the instruments agree reasonably well in terms of the seasonal cycle and magnitude of cloud particle/cloud residual concentrations (Fig. 2a). A 2D histogram of ~~corrected~~ the cloud particle concentrations versus cloud residual concentrations (Fig. 2b) shows that most of the data points lie on or around the 1:1 line. ~~An orthogonal linear regression~~ 65% of the data lie within a factor of 2, 88% within a factor of 5, and 92% within a factor of 10 from the 1:1 line. A linear orthogonal distance regression (ODR) of cloud residual versus cloud particle number concentrations (Fig. 2b) returns a slope of ~~0.97, an offset of 4.9 cm^{-3} and an~~ 1.14 and a coefficient of determination, R^2 ~~of 0.47~~, of 0.61. However, there is ~~a substantial amount of~~ some scatter. Most notably, there is a ~~cloud~~ group of data points below the 1:10 line ($\sim$7–8% of the data) that seems to be associated with colder temperatures at the sampling site (Fig. 2c). Due to this temperature dependent behaviour, we will derive and discuss the correction factors for warm and cold clouds separately.

~~Temperatures~~

3.1.1 Warm clouds

In Fig. 2, we corrected the cloud particle concentrations for the GCVI transmission efficiency. However, to be able to compare the cloud residual measurements to the aerosol particle measurements from the whole-air inlet, we need to apply the correction in the other direction. The integrated transmission efficiency of the GCVI inlet was estimated by comparing the integrated cloud particle number concentrations with and without taking into account the size-dependent transmission efficiency of Shingler et al. (2012). Figure 3 shows the results (for temperatures > 0°C) when integrating over the entire cloud particle population and when integrating only above the cut-size diameter of the GCVI. Above the GCVI droplet/crystal cut-size (red histogram in Fig. 3a), the distribution is symmetrical and relatively narrow, and it shows that approximately half (mean $\pm$ std ratio of 0.5 ± 0.04) of the total cloud particles were sampled. Figure 3b shows the corresponding 2D histogram, together with an ODR fit which returned a best fit slope of 0.51.

For warm (liquid) clouds, the sampling efficiency could also be estimated by scaling the cloud residual size distribution to the total particle size distribution under the assumption that all accumulation mode particles activate into cloud droplets.

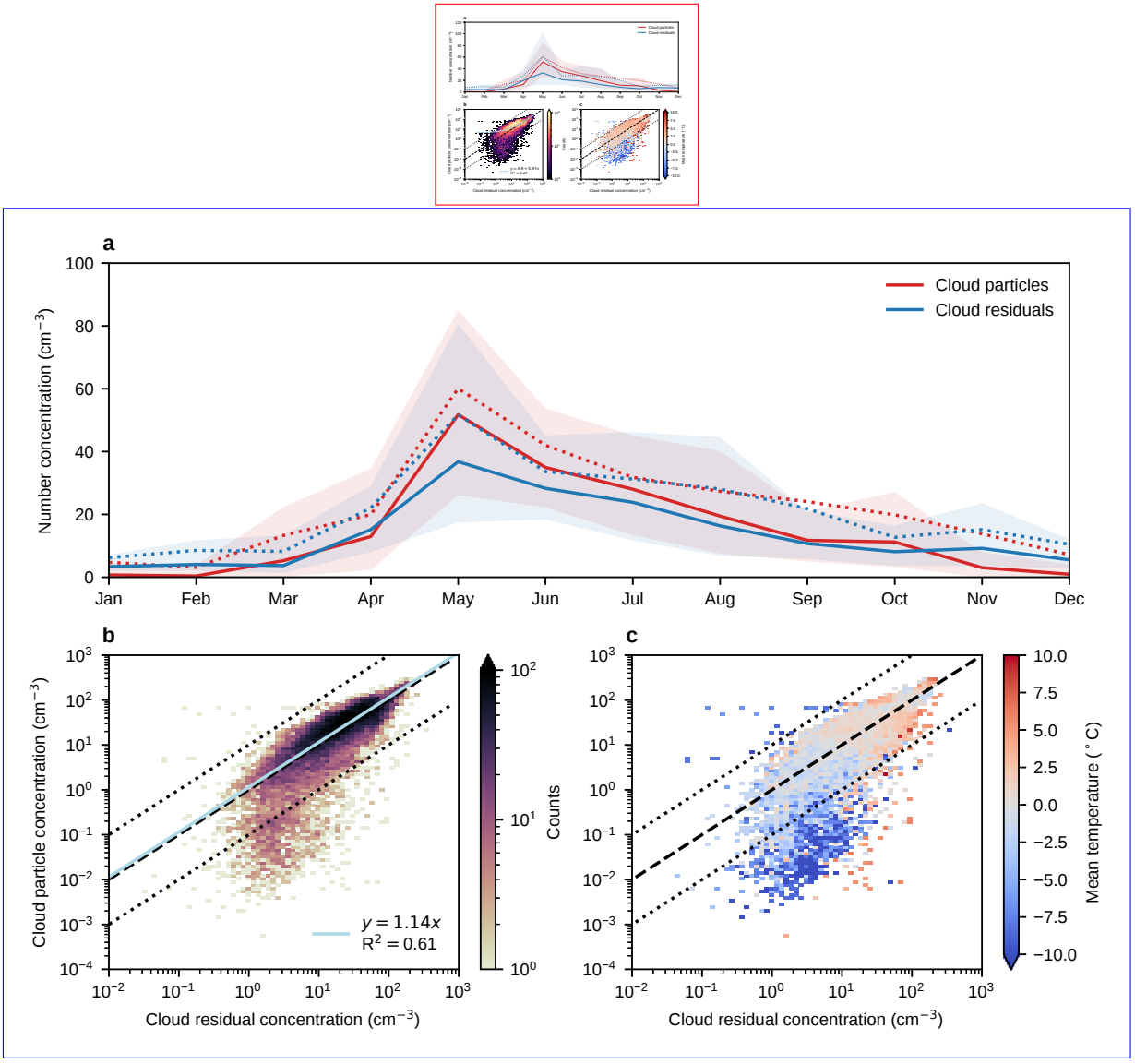

Figure 2. ~~Comparison of cloud residual number concentrations and ambient cloud particle concentrations.~~ Comparison of cloud residual and cloud particle number concentrations. **a** Monthly averages of cloud residual number concentrations as measured behind the GCVI (blue) and cloud particle number concentrations ~~as measured by~~ derived from the FM-120 fog monitor measurements (red). Solid and dotted lines show ~~mean and~~ median and mean values, respectively, and shaded areas indicate the 25th to 75th percentile ranges. **b** Density scatterplot of cloud residual versus cloud particle number concentrations, including an orthogonal distance linear regression (grey line). **c** The same as b, but colourcoded by the average temperature instead of the data point density. In b and c, the black dashed line represents the 1:1 line and the dotted lines represent 10:1 and 1:10 lines. The transmission efficiency and cut-size of the GCVI inlet (Shingler et al., 2012) has been included in the calculation of the cloud particle number concentration in all panels.

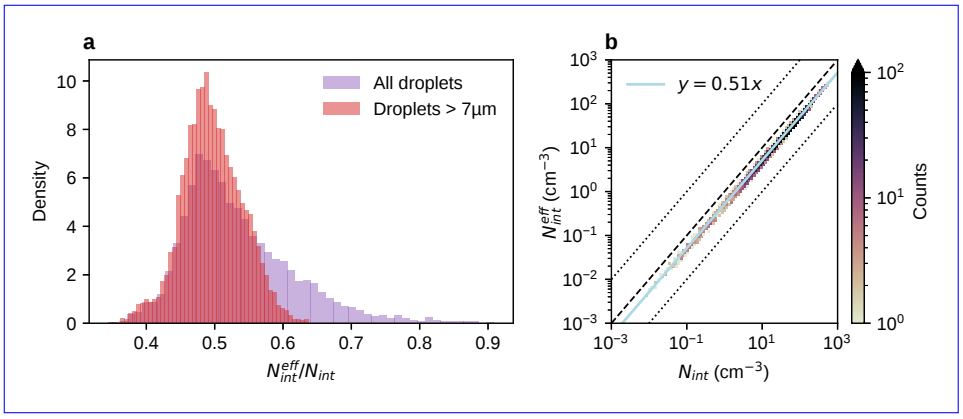

Figure 3. Cloud particle number concentrations with and without taking the GCVI sampling efficiency into account. Comparison of fog monitor total integrated cloud particle number concentration after correcting for the GCVI transmission efficiency (Shingler et al., 2020) ($N_{\mathrm{int}}^{\mathrm{eff}}$) divided by the total integrated cloud particle number concentration without correction (N_{int}), for temperatures $> 0°$C. **a** shows histograms of the ratio for the whole cloud particle size range (purple) and when integrating the cloud particle distribution above the GCVI size cut-off (red). **b** shows a density scatter plot of N_{int} versus $N_{\mathrm{int}}^{\mathrm{eff}}$ for the points from the red histogram, together with an ODR best fit slope.

We compared cloud residual and total particle concentrations (30 min mean values) integrated above different accumulation mode threshold diameters, D_{cut}, in the range 41–505 nm. Figure 4a shows an example scatter plot for $D_{\mathrm{cut}} = 123$ nm, with the corresponding ODR fit parameters (grey data points correspond to cold clouds and are not included in the fit). Figure 4b–c shows ODR best fit slopes and coefficients of determination for all the different D_{cut} diameters (results from cold cloud data are included for completeness, but not used since we then cannot assume liquid droplet activation).

Above approximately 100 nm, the slope plateaus around 0.46. Given that this method does not take into account the cloud particle cut-size of the GCVI inlet, we can expect this estimate of the transmission efficiency to be a little lower than the one derived from the fog monitor data (0.51). A value of 0.46 is just within the standard deviation of the ratio from Fig. 3, so these two independent methods agree remarkably well. To be conservative, and to be consistent with the cold clouds (next subsection), we have corrected all (warm) cloud residual size distributions and concentrations by a factor of 2 (from Sect. 3.2.1 onward) assuming cloud residual size and cloud particle size are not correlated.

3.1.2 Cold clouds

At temperatures below 0°C ~~could indicate the presence of~~, things may become more complicated since we could be sampling not only droplets but also ice crystals (e.g. in ice or mixed-phase clouds). The same analysis as in Fig. 3 is shown for cold clouds in Fig. 5. The ratio distribution is almost identical to the warm case, with a mean±std of 0.5 ± 0.05. The ODR best fit slope of 0.53 is also very similar to the warm case (0.51). The shape of the cloud particle number size distribution was relatively constant during the sampling period (cf. Fig. S4), and the majority of the sampled cloud particles at temperatures $\leq 0°$C are likely to be

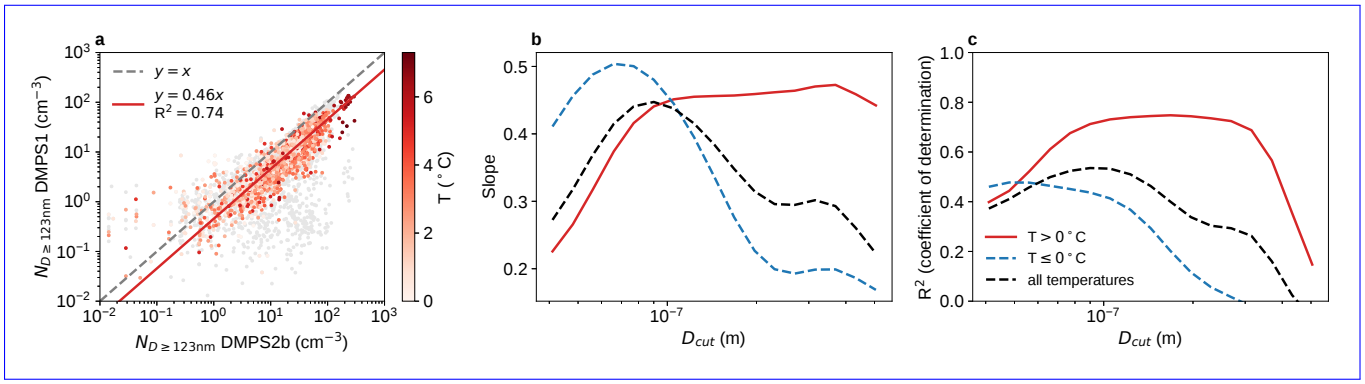

Figure 4. **Comparison of total and residual accumulation mode particle concentrations.** **a** scatterplot of accumulation mode (here: diameter $>$ 123 nm) concentrations from DMPS 2b and DMPS 1 for temperatures $>0°$C, together with an ODR best fit slope. Temperatures $\leq 0°$C are shown as grey points but are not included in the fit. **b** and **c** show ODR best fit slopes and corresponding coefficients of determination, respectively, for different definitions of the accumulation mode. The red lines show these parameters for temperatures $>0°$C, and the dashed lines show the corresponding values for temperatures $\leq 0°$C (blue) and all data (black), included for completeness.

supercooled droplets, which can explain the similarity in behaviour between the warm and cold cases. Because the behaviour is so similar to the warm case, we have also corrected all cold cloud residual size distributions and concentrations by a factor of 2 (from Sect. 3.2.1 onward) assuming cloud residual size and cloud particle size are not correlated. However, Fig. 2b–c clearly showed that there are a few cases, mainly at very cold temperatures, where the assumed transmission efficiency (cf. Fig. S4) cannot reconcile the cloud particle and cloud residual concentrations measured. This will be discussed below.

3.1.3 Outliers at cold temperatures

Figure 2b showed that there were two groups of data points: one with good agreement between cloud residual and cloud particle concentrations, and one where the cloud residual concentrations exceeded the cloud particle concentrations by one to two orders of magnitude. The second group is associated with very low cloud particle concentrations ($1 \pm 3\,\mathrm{cm}^{-3}$, total concentrations without cut-size and Shingler et al. (2012) correction) and the cloud particles are also fairly large in size ($11 \pm 4\,\mu$m effective radius). While this group is only a small minority of the data ($\sim$7–8%), its correlation with cold temperatures (Fig. 2c) means it warrants further investigation.

A discrepancy like this can have two basic causes:

a) The apparent concentration difference does not reflect the true difference but is a result of varying sampling efficiencies/issues of the FM-120 and the GCVI, or

b) The concentration difference is true, but can be a result of either real physical processes in the atmosphere or of spurious measurements caused by sampling artefacts

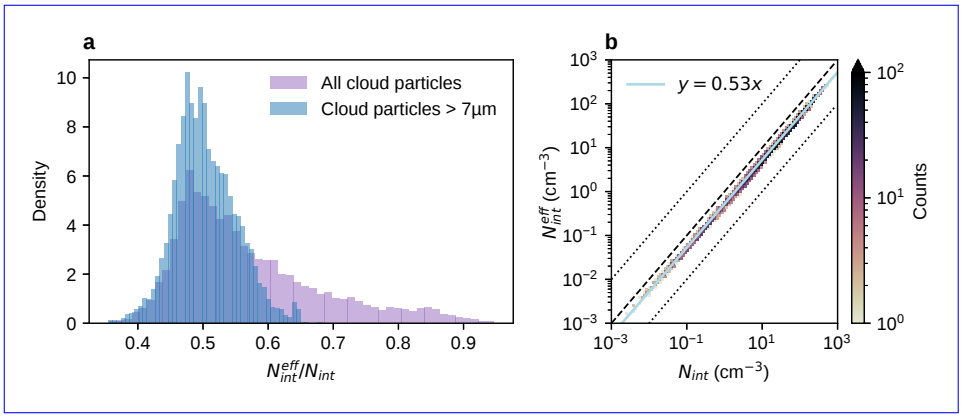

Figure 5. Cloud particle number concentrations with and without taking the GCVI sampling efficiency into account. Comparison of fog monitor total integrated cloud particle number concentration after correcting for the GCVI transmission efficiency (Shingler et al., 2020) ($N_{\mathrm{int}}^{\mathrm{eff}}$) divided by the total integrated cloud particle number concentration without correction (N_{int}), for temperatures $\leq 0^{\circ}$C. **a** shows histograms of the ratio for the whole cloud particle size range (purple) and when integrating the cloud particle distribution above the GCVI size cut-off (blue). **b** shows a density scatter plot of N_{int} versus $N_{\mathrm{int}}^{\mathrm{eff}}$ for the points from the blue histogram, together with an ODR best fit slope.

Determining the cause is not a trivial task. Both the GCVI and the FM-120 were calibrated using spherical standard particles, which makes the comparison especially difficult for cases when ice crystals are sampled. The true transmission efficiency of the GCVI inlet is going to be different for non-spherical particles, i.e. ice crystals, which are not accurately represented by glass beads. In addition, the concept of size becomes ambiguous when the sampled particles are not spherical, especially since the two instruments deal with different types of size. The optical size reported by the FM-120 is not necessarily the same as the Stokes equivalent size that determines how a crystal behaves inside the GCVI inlet, which means that the transmission efficiency we apply could be incorrect. ~~For non-spherical ice crystals, the sizing uncertainties in the fog monitor can be larger than those associated with Mie theory (Baumgardner et al., 2017), and as a result under- or oversizing of crystals can occur. This could also affect the concentration comparison.~~ However, the points below the 1:10 line in Fig. 2~~(~ 7–8% of the data)~~ b–c still remain below the 1:1 line even if we compare the cloud residual concentration to the total, uncorrected cloud particle concentration (not shown), which suggests that something other than errors in the assumed transmission efficiency is causing the difference.

~~Riming or impaction scavenging of interstitial aerosol particles onto an ice crystal could result in more than one cloud residual emerging from the crystal as it dries inside the GCVI inlet (e.g., Mertes et al., 2007; Santachiara et al., 2018). Similarly, an INP could break or eject material during the freezing process (e.g., Lauber et al., 2018) which could also result in more than one residual particle per crystal. These processes could make cloud residual concentrations exceed cloud particle concentrations; however, the concentrations in Fig. 2 sometimes differ by almost two orders of magnitude, and it is unlikely that the aforementioned processes could account for the full difference (Santachiara et al., 2018).~~

~~Measurement artefacts during in-situ sampling of cloud droplets and ice crystals are a common and complex challenge (Baumgardner et al., 2017) contributing to both overestimation and underestimation of cloud residual (or cloud particle) concentration measurements (Pekour and Cziczo, 2011; Spiegel et al., 2012; Shingler et al., 2012). Particle capture by wake effects in the GCVI inlet is a possible explanation for cloud residual concentrations exceeding cloud particle concentrations; however, even at cloud particle and interstitial aerosol particle concentrations at the upper end of our observed values, only 1 % of the measured cloud residuals is estimated to be a potential artefact (Pekour and Cziczo, 2011). Thus, this effect is likely not the major cause of the disparity between cloud residual and cloud particle concentrations that sometimes occurs. Droplet or ice crystal shattering is another potential source of small particles within the acceleration and deceleration zones of the GCVI, and this could also cause an overestimation of the cloud residual number concentration. Intuitively, one would expect shattering events to produce large amounts of particles, yet the largest relative differences between cloud particle and cloud residual concentrations mainly occur at very low particle concentrations. Nevertheless, the apparent correlation with cold temperatures (Fig. 2c) means that ice crystal shattering inside the GCVI inlet cannot be fully ruled out.~~

A comparison of the measured visibility and the visibility calculated from the FM-120 data shows a reasonable agreement for the majority of data points, but again, as in Fig. 2c, there is a group of data points at predominantly cold temperatures where the agreement is much worse (Fig. S5a–b in supplementary material). The visibility was calculated using the Koschmieder formula (Seinfeld and Pandis, 2016), the measured cloud particle size distribution of the FM-120 and Mie theory (Python package *PyMieScatt* (v. 1.7.5); Sumlin et al., 2018) by assuming spherical particles with the refractive index of water (1.33) and a wavelength of 880 nm (of the visibility sensor). We have already suggested that the assumption of spherical particles might not hold at cold temperatures, which could explain the differences in Fig. S5. However, the calculated visibility is sometimes several orders of magnitude higher than the measured one, and it ~~seems~~ is unlikely that non-sphericity would cause such large differences ~~.~~ given that the overall sizing uncertainty from optical particle spectrometers only increases from ±20 % to around ±30 % for non-spherical particles (Baumgardner et al., 2017; Borrmann et al., 2000). One possibility is that the differences in visibility are caused by a loss of detected cloud particles within the FM-120 (e.g. turbulent deposition inside the contraction of the inlet) or that cloud particles (e.g. larger ice crystals or snow flakes) are larger than the last channel of the FM-120. However, in the latter case, the cloud particles are also likely to be too large to be sampled by the GCVI.

The FM-120 as well as the GCVI inlet sampling efficiency can also be affected by the wind speed and direction (Spiegel et al., 2012), but this is not something we can easily correct for. The FM-120 faces south, i.e. into the prevailing winds at Zeppelin Observatory. The station experiences northerly winds approximately one third of the time during cloud events (see Fig. S1), and one might expect this to reduce the sampling efficiency of the FM-120. However, heatmaps similar to Fig. 2c for wind speed, updraft, and wind direction indicate no obvious correlation between wind parameters and deviations of concentrations from the 1:1 line (see Fig. S6). Interestingly, the two data groups show similar internal gradients of wind speed and updraft velocity (Fig. S6a–b), which may suggest that the gradients are caused by the same physical process(es) but the second data group is just shifted by e.g. undercounting in the FM-120. In general, no clear influence of the prevailing wind direction in the comparison of residual and cloud particle concentration can be observed (see Fig. S7). Local effects of blowing snow could also affect the measured visibility, but there is no overall correlation between wind speed and differences in cloud particle and cloud residual

concentrations (see Fig. S6). For both cases, one should also take into account that high wind speeds are only rarely observed at Zeppelin Observatory (the median wind speed is approximately 3 ms^{-1}, see Fig. S1).

The presence of precipitating particles may cause the measured visibility to be higher than the calculated one; however, precipitating particle concentrations at Zeppelin Observatory have previously been found to be mostly lower than 0.3 cm^{-3} (Koike et al., 2019). Hence, while the presence of such particles could explain the differences in visibility, they ~~do not~~ can only partially explain the differences in concentration in Fig. 2~~. Local effects of blowing snow could also affect the measured visibility, but there is no apparent correlation between wind speed and differences in cloud particle and cloud residual concentrations (see Fig. S6 in supplementary material). Thus, we are left with the possibility that the differences in visibility are caused by a loss of detected cloud particles within the FM-120. It could be that ice crystals are more susceptible to losses due to turbulent deposition inside the contraction of the inlet (Spiegel et al., 2012).~~

~~Recalculating the visibility after scaling up the cloud particle size distributions to the measured cloud residual concentrations (i.e.~~b unless significant ice crystal shattering (e.g.~~ multiplying each cloud particlesize distribution by the ratio of the total cloud residual concentration and the cloud particle concentration integrated above the GCVI cut-size) significantly improves the agreement with the measured visibility (see Fig.~~from snow flakes) occurs in the GCVI wind tunnel. At air speeds of 100 ~~S5c–d in supplementary material). Note that the concentrationswere only scaled up, not down, since the total cloud particle concentrations could be higher than the cloud residual concentrations due to~~ ms^{-1}, the critical diameter above which droplets may shatter into fragments is $\sim 76\,\mu$m (Twohy et al., 2003). Precipitating particles can exceed this size, and could thus produce fragments that are sampled if they are larger than the aerodynamic cut-size of the ~~GCVI inlet. The improvement in the visibility comparison after scaling the cloud particle concentrations points towards an undercounting of cloud particles in the FM-120 for parts of the period with colder temperatures~~GCVI. However, it is likely that many of the fragments would not be aligned with the streamlines and therefore would not enter the GCVI.

Measurement artefacts during in-situ sampling of cloud droplets and ice crystals are a common and complex challenge (Baumgardner et al., 2017) contributing to both overestimation and underestimation of cloud residual (or cloud particle) concentration measurements (Pekour and Cziczo, 2011; Spiegel et al., 2012; Shingler et al., 2012). Particle capture by wake effects in the GCVI inlet is a possible explanation for cloud residual concentrations exceeding cloud particle concentrations; however, even at cloud particle and interstitial aerosol particle concentrations at the upper end of our observed values, only 1 % of the measured cloud residuals is estimated to be a potential artefact (Pekour and Cziczo, 2011). One would also expect to see the influence of the wake effect in the summer months but in our case the disagreement is mainly seen in winter months with generally low concentrations. Thus, ~~it cannot be ruled out that the discrepancy between the measured and calculated visibilities, and by extension the discrepancy~~ this effect is likely not the major cause of the disparity between cloud residual and cloud particle concentrations ~~, is partly due to the FM-120 undercounting the ambient ice crystals~~that was observed.

~~As we have seen, interpreting cloud residual data is a non-trivial task. There are many processes, both natural and instrument related, that can influence the measured concentrations of both cloud residuals and cloud particles. We cannot definitively say which sources of error are affecting which instrument, and therefore we will not discard any data at this stage. Still, a~~

more detailed comparison with respect to temperature and wind parameters – also including data from the whole-air inlet – is warranted, and this is done in the next subsections.

In Fig. 2, we corrected the cloud particle concentrations for the GCVI transmission efficiency. However, to be able to compare our cloud residual measurements to the aerosol particle measurements from the whole-air inlet, we need to apply the correction in the other direction. The integrated transmission efficiency of the GCVI inlet was estimated by calculating the ratio of the integrated cloud particle number concentrations with and without taking into account the size-dependent transmission efficiency of Shingler et al. (2012).Figure **??** shows the results when integrating over the entire cloud particle population and when integrating only above the cut-size diameter of the GCVI. Above the GCVI droplet/crystal cut-size (red histogram in Fig. **??**a), the distribution is symmetrical and relatively narrow, and it shows that approximately half (mean ± std ratio of 0.5 ± 0.05) of Droplet or ice crystal shattering is another potential source of small particles. Shattering could either happen in the wind tunnel or after the stagnation plane within the CVI inlet, and this could also cause an overestimation of the cloud residual number concentration. If the particles were to shatter after the stagnation plane, this should be clearly seen as spikes in the cloud residual concentrations measured by the total CPC, and this was not observed. Regarding shattering in the wind tunnel, as stated above, the total cloud particles were sampled. The mode of the distribution shows very little variation between seasons (Fig.cloud particles need to exceed a certain size for this process to be likely. While this can happen when there is precipitation, it needs to be borne in mind that in most non-precipitating clouds, the concentration of large cloud particles is much lower than the total cloud particle concentration, and therefore particles that do shatter may need to far exceed the critical break-up diameter to produce enough fragments to significantly increase the measured cloud residual concentration (Twohy et al., 2003). If the concentration differences we observe are caused by shattering artefacts, then, the magnitude of the difference suggests that precipitating particles (e.g. **??**b). We have therefore corrected all snow) are a more likely cause than large cloud droplets or ice crystals.

Riming or impaction scavenging of interstitial aerosol particles onto an ice crystal may be able to result in more than one cloud residual emerging from the crystal as it dries inside the GCVI inlet. If more than one residual could be released through this process without the need for crystal break-up, it could be an alternative explanation that is consistent with the difference in cloud residual and cloud particle number concentrations. However, this is speculative since no strong experimental evidence exists on how rimed particles would behave inside the CVI flow regime.

In summary, the relatively small amount of outliers discussed in this section could be a result of snow or ice crystal shattering, but there are also possible alternative physical explanations. In the following sections, we continue to segregate the cloud residual data based on temperature and will later use cluster analysis of the shape of the cloud residual size distributions and concentrations by a factor of 2 assuming cloud residual size and cloud particle size are not correlated. An individual correction factor for each data point would, in theory, be possible, but only for the points where we have overlapping cloud particle and cloud residual data. Thus, for the sake of consistency, we use a constant correction factorfurther investigate the potential role of artefacts and ice/snow within the clouds we observed.

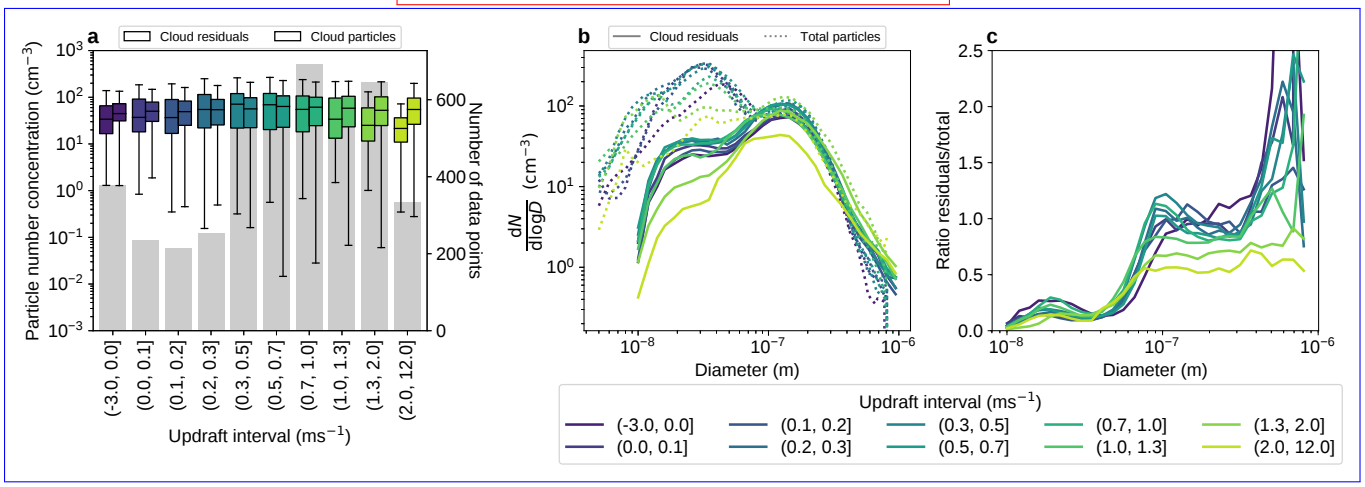

Figure 6. In-cloud data binned by updraft. a Box plot of cloud residual (solid) and cloud particle (hatched) number concentrations for different updraft intervals (see legend). The whiskers extend to the farthest points that are within 1.5 times the interquartile range from the nearest quartile. Points that fall outside the whiskers are not shown. The grey bars in the background indicate the number of cloud residual data points (right y-axis) per updraft bin. **b** Mean particle number size distributions of cloud residuals (solid) and total particles (dotted) for different updraft intervals (see legend). **c** Ratio of the size distributions in b, i.e. cloud residual concentrations divided by total particle concentrations.

3.2 Two years of cloud residual size distributions

3.2.1 Warm clouds

In liquid droplet activation, the updraft velocity is an important driver (alongside the aerosol particle size distribution and the particle composition) since it controls the supersaturation. We will therefore study how the cloud residual size distribution and number concentration varies with varying updraft velocity. Because Zeppelin Observatory is a mountain site, a closer look at the updraft is also warranted to investigate potential orographic effects.

3.3 ~~Comparison with respect to ambient (acoustic) temperature~~

Figure ~~7~~ 6 shows concurrent cloud particle, cloud residual and total aerosol particle data binned by ~~ambient temperature (acoustic temperature recorded by the anemometer). Note that the acoustic temperature was, on average, 3.4°C higher than the actual air temperature (see Sect. 2.3.3) at Zeppelin Observatory during our measurement period~~updraft velocity. Panel a

shows box plots of cloud residual concentrations (corrected by the factor 2, see above) and cloud particle concentrations (now without correction, but still integrated only above the GCVI cut-size). The concentrations ~~agree well down to about -2 to -4°C, where the cloud particle concentrations drop below~~ generally agree well, but there seems to be a tendency for the cloud residual ~~concentrations. These bins contain relatively few data points (bar plot in Fig.~~ number concentrations to be underestimated at higher updraft, starting approximately above 1–1.3 ~~7a), but they follow the general trend of decreasing cloud particle~~/ms^{-1}. Panels b and c show a similar pattern, with cloud residual concentrations ~~with decreasing temperature.~~

~~**In-cloud data binned by temperature. a** Box plot of cloud residual (solid) and cloud particle (hatched) number concentrations for different temperature intervals (see legend). Note that the temperature is the acoustic temperature measured by an anemometer. The grey bars in the background indicate the number of cloud residual data points (right y-axis) per temperature bin. **b** Mean particle number size distributions of cloud residuals (solid) and total particles (dotted) for different temperature intervals (see legend). **c** Ratio of the size distributions in b, i.e. cloud residual concentrations divided by total particle concentrations.~~

~~Figure~~decreasing in the last two or three updraft bins. This pattern is not observed in the total aerosol particles, except for in the highest updraft bin (Fig. ~~7b shows the mean cloud residual and total aerosol particle size distributions for the same temperature bins. Both the Aitken and the accumulation mode are present in both cloud residuals and total particles, but the total particle size distributions generally show higher particle concentrations, particularly of Aitken mode particles.~~ 6b).

Figure ~~7~~6c shows the ratio between the distributions in panel b, i.e. cloud residual concentrations divided by total particle concentrations. ~~At temperatures above approximately -2°C, the resulting curves are sigmoidal~~ The curves are more or less sigmoidal, like typical CCN-activated particle fraction curves. ~~During cloud events at these temperatures, the figure shows that most of the total aerosol particles larger ~100 nm are in fact cloud residuals. The apparent~~ The $D_{50\%}$, defined as the diameter where the ratio is around 0.5, ~~decreases with decreasing temperature, which could be related to the general decrease in particle concentrations with temperature seen in the other two panels of the figure, since lower overall particle concentrations could allow smaller particles to activate (assuming liquid droplet activation without size-dependent chemical composition if the meteorological conditions are the same)~~ranges between approximately 58 and 78 nm. In Fig. 6b, clear changes in the cloud residual size distributions (i.e. shift towards smaller activation diameters) are only seen for updrafts above around 2 ms^{-1}, which is a relatively small subset of the observations. Thus, the stated $D_{50\%}$ range is unlikely to be influenced by orographic effects.

Most of the curves level out around a ratio of 1, indicating that most of the total aerosol particles larger than ~100 nm are in fact CCN. Despite all the uncertainties and assumptions being made (e.g. ~~CVI~~ GCVI sampling efficiency and enrichment factor), it is encouraging to see that overall ratios are in the range of expected values giving further faith in our observations. ~~Ratios~~ The ratios are occasionally above 1 ~~can be explained by the uncertainties in sampling efficiency and enrichment factor of the CVI system (see Sect. 3.1 above) and small uncertainties in sizing, concentration and losses of the DMPS.~~

~~At temperatures below -2°C (approximately 16 % of this subset of data), however,~~ which, at the upper end of the ~~curves in Fig. 7c look very different. Instead of an S-shape, the curves are relatively flatter with a maximum appearing at lower sizes, with the coldest temperature bins even showing a peak below ~20 nm particle diameter. This implies that accumulation mode particles have not acted as cloud residuals, while now an increased contribution of Aitken mode particles served as cloud~~

seeds. These clouds most likely contain ice particles and the question arises if the small particles could potentially particle size range, could be caused by sampling artefacts inside the CVI sampling line (see Sect. 3.1 above) or if real physical atmospheric process is underlying this observation.While artefacts cannot be completely ruled out (see Sectsmall number statistics (i.e. 3.1), it should be noted that approximately the same size modes are present in the whole-air inlet (Fig. 14), and there is no reason to expect that any potential droplet or crystal shattering in this inlet should produce the same size particles as shattering in the GCVI inlet. The shape of the ratio curves in Fig. 7c below -2°C indicate the participation of ice in cloud formation as will be discussed in Sect. **??** within a cluster analysis of the residual size distributions.

3.3 Comparison with respect to updraft

ratios of small numbers). 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.

The FM-120 as well as the GCVI inlet sampling efficiency can also be affected by the wind speed and direction (Spiegel et al., 2012), but this is not something we can easily correct for. However, heatmaps similar to Fig. 2c for wind speed, updraft, and wind direction indicate no obvious correlation between wind parameters and deviations of concentrations from the 1:1 line (see Fig. S6). One should also take into account that high wind speeds are only rarely observed at Zeppelin Observatory (the median wind speed is approximately 3 ms^{-1}), which can be seen in Fig. S1. Nevertheless, since Zeppelin Observatory is a mountain site, a closer look at the updraft is warranted to investigate potential orographic effects.

In-cloud data binned by updraft. a Box plot of cloud residual (solid) and cloud particle (hatched) number concentrations for different updraft intervals (see legend). The grey bars in the background indicate the number of cloud residual data points (right y-axis) per updraft bin. **b** Mean particle number size distributions of cloud residuals (solid) and total particles (dotted) for different updraft intervals (see legend). **c** Ratio of the size distributions in b, i.e. cloud residual concentrations divided by total particle concentrations.

Figure 6 shows concurrent cloud particle, cloud residual and total aerosol particle data, this time binned by updraft velocity instead of acoustic temperature. The box plots in the first panel show that the cloud residual and cloud particle number concentrations generally agree well, but there seems to be a tendency for the cloud residual number concentrations to be underestimated at higher updraft, starting approximately above 1 ms^{-1}. Panels b and c show a similar pattern, with cloud residual concentrations decreasing in the last four updraft bins. This pattern is not observed in the total aerosol particles, except for in the highest updraft bin (Fig. 6b). The curves in Fig. 6c systematically level out at lower ratios with higher updrafts (for the last four two or three bins), which could either mean that not all accumulation mode particles are cloud residuals CCN under these conditions, or that the GCVI inlet fails to sample all cloud particles at high updrafts. Taken together with the previous panels, it seems likely that the GCVI inlet sampling efficiency is negatively affected by high updraft velocities (or indeed high wind speeds in general, as these parameters tend to be correlated at Zeppelin Observatory). The sampling efficiency of the FM-120 fog monitor can in theory also be adversely affected by high wind speeds (Spiegel et al., 2012), but this seems to happen to a lesser extent than for the GCVI inlet based on Fig. 6a. One should bear in mind that the wind speeds and updrafts are generally lower near the FM-120 as it is positioned at a lower altitude than the GCVI inlet (∼5 m below).

3.0.1 Cold clouds

~~During cloud events within our measurement period, typical cloud residual number concentrations ranged between 10 and 62~~Figure ~~cm^{-3} (25^{th} and 75^{th} percentiles), with a median of 25 cm^{-3} (mean $\pm$ standard deviation: 50 ± 66 cm^{-3}). Total concentrationsof particles suspended in the air (diameters 10–809~~7 shows concurrent cloud particle, cloud residual and total aerosol particle data, this time binned by ambient temperature instead of updraft velocity. The box plots in the first panel show that the cloud residual and cloud particle number concentrations agree well down to about -4 to -6°C, where the cloud particle concentrations drop below the cloud residual concentrations. These bins contain relatively few data points (bar plot in Fig. ~~nm)during these cloud events were generally higher, ranging up to 163 cm^{-3} (75^{th} percentile)with a median of 70 cm^{-3} (mean $\pm$ standard deviation: 145 ± 235 cm^{-3}) .~~ 7a), but they follow the general trend of decreasing cloud particle/cloud residual concentrations with decreasing temperature.

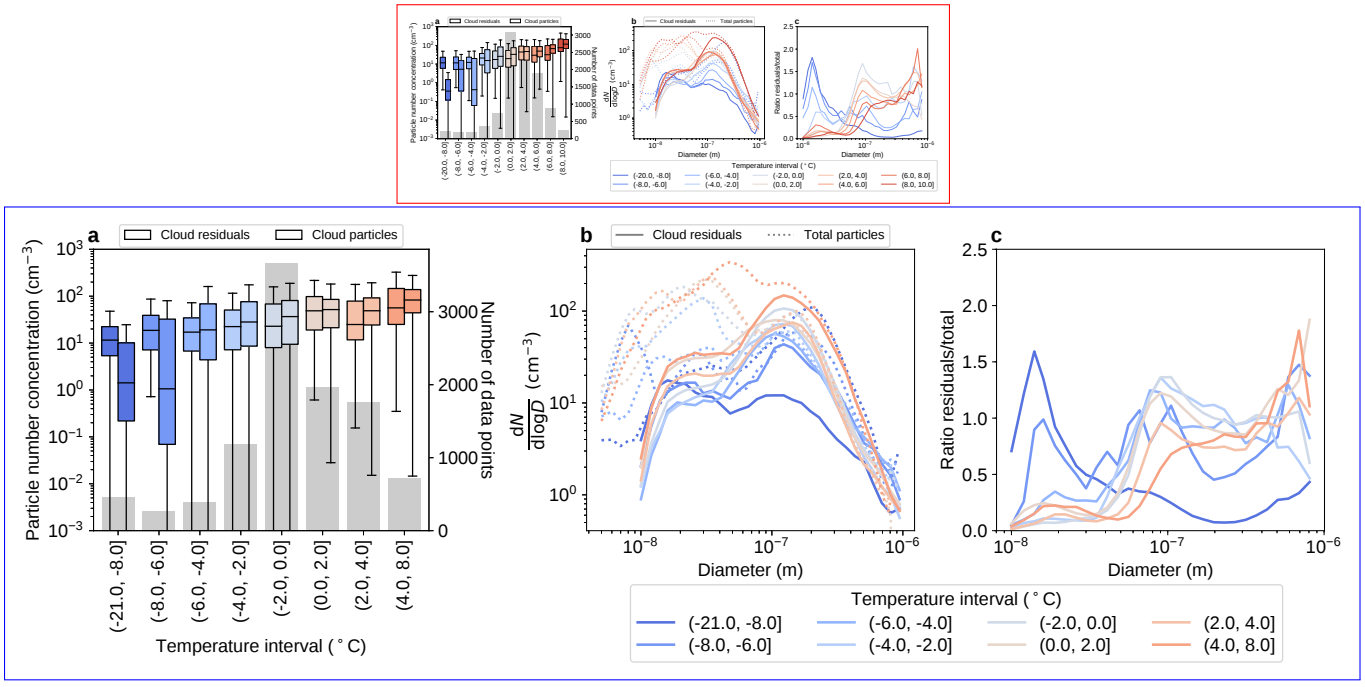

Figure 7. In-cloud data binned by temperature. a Box plot of cloud residual (solid) and cloud particle (hatched) number concentrations for different temperature intervals (see legend). The whiskers extend to the farthest points that are within 1.5 times the interquartile range from the nearest quartile. Points that fall outside the whiskers are not shown. The grey bars in the background indicate the number of cloud residual data points (right y-axis) per temperature bin. **a** Mean particle number size distributions of cloud residuals (solid) and total particles (dotted) for different temperature intervals (see legend). **c** Ratio of the size distributions in b, i.e. cloud residual concentrations divided by total particle concentrations.

Figure 7b shows the mean cloud residual and total aerosol particle size distributions for the same temperature bins. Both the Aitken and the accumulation mode are present in both cloud residuals and total particles, but the total particle size distributions generally show higher particle concentrations, particularly of Aitken mode particles. At temperatures above approximately -4°C, the ratio curves in Fig. 7c have a similar shape as for the pure liquid clouds (cf. Fig. 6). This could be an indication that in the temperature range -4–0°C we are mostly sampling supercooled droplets. The apparent $D_{50\%}$ decreases with decreasing temperature, which indicates an increase in cloud supersaturation with decreasing temperature. If the meteorological conditions are otherwise the same, this could be caused by an increase in updraft velocity or by a decrease in particle concentration (less competition for water vapour allows smaller particles to be activated). The latter is consistent with the general decrease in particle concentrations with temperature seen in the first two panels of Fig. 6.

At temperatures below -4°C (approximately 11 % of this subset of data), however, the curves in Fig. 7c look very different. Instead of an S-shape, the curves are relatively flatter with a maximum appearing at lower sizes, with the coldest temperature bins even showing a peak below ∼20 nm particle diameter. Assuming that the measured cloud residuals directly correspond to CCN or INP, this behaviour implies that many of the ambient accumulation mode particles have not activated, while now an increased contribution of Aitken mode particles served as cloud seeds. These clouds most likely contain ice particles and the question arises if the small particles could potentially be caused by sampling artefacts inside the GCVI system (see Sect. 3.1.3 above) or if a real physical atmospheric process is underlying this observation. This will be further discussed in Sect. 3.0.2.

3.0.2 Annual cycle

Our observations cover two full years and allow us to study seasonality effects, which are expected for this remote Arctic site, where aerosol properties follow recurring patterns (e.g., Tunved et al., 2013) that will also influence the seasonality of cloud residual properties. We will follow two approaches to study the seasonality of cloud residuals: (a) grouping the observations in liquid and mixed-phase clouds and (b) using a cluster analysis of the shape of the cloud residual size distributions. The latter approach will allow us to identify and quantify the impact of potential measurement artefacts or the contribution of ice which will be discussed in detail below.

Separation between liquid and mixed-phase clouds

Figure 8 shows concurrent monthly averages of total particle and cloud residual number concentrations integrated above 20 nm diameter. Panel a shows all data, whereas panels b and c show data for ~~updraft velocities below and above 1 ms^{-1}~~ temperatures above and below $-4°C$, respectively. This boundary was chosen ~~partly based on~~ based on the different behaviours seen in Fig. ~~6, but also because 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 orographic effects at the station~~7c, where cloud residual size distributions below $-4°C$ are more likely to be influenced by ice, while cloud residual distributions above $-4°C$ are mostly liquid clouds. The bar charts next to each panel indicate the number of data points per month.

The observed total particle number concentrations follow the typical seasonal cycle of Arctic aerosol. We recognise the characteristic maxima in number concentration due to Arctic haze in spring and new particle formation in summer, and the

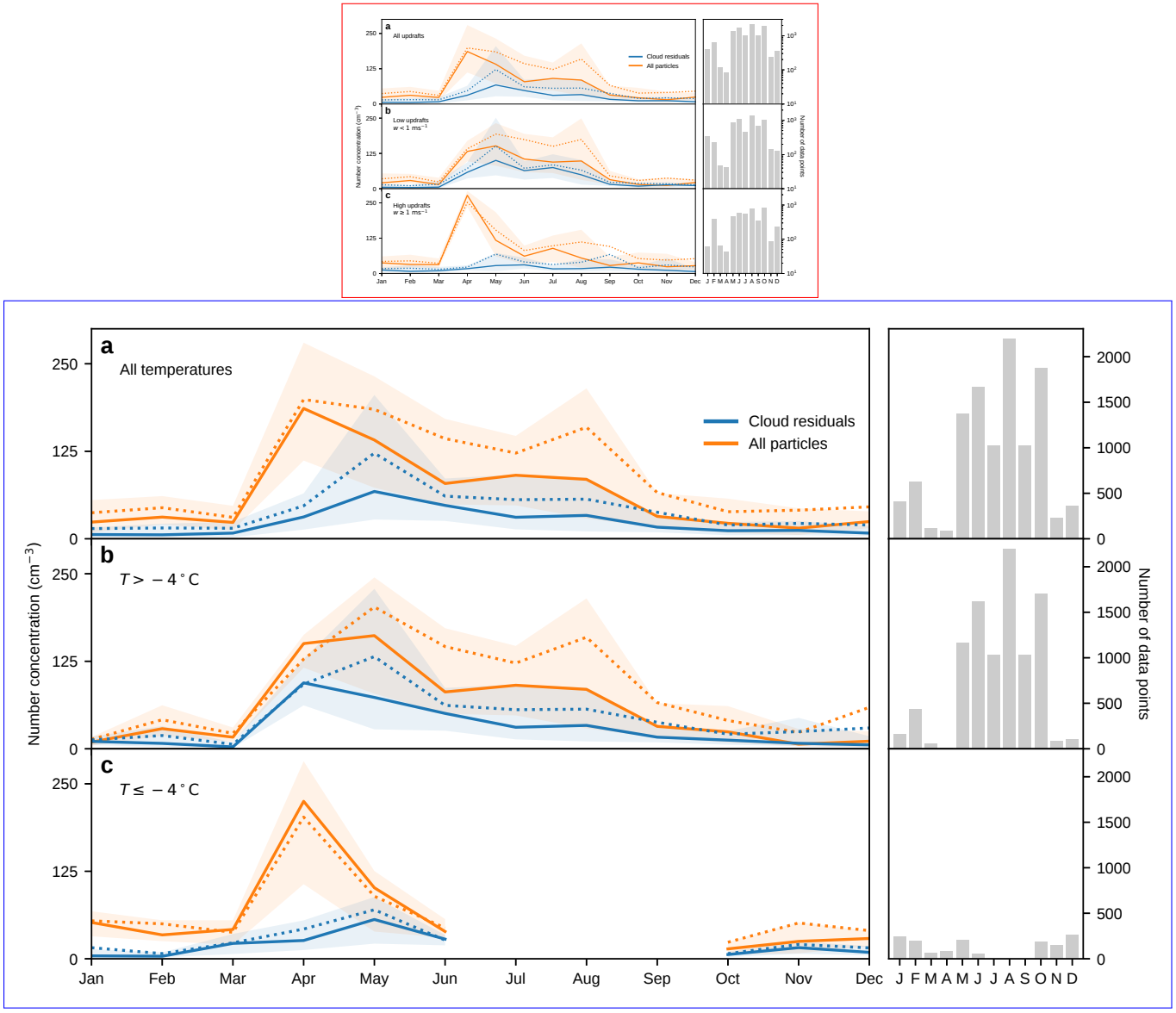

Figure 8. Annual cycle of total and cloud residual number concentrations. Monthly averages of total (orange) and residual (blue) particle number concentrations integrated above 20 nm, measured at Zeppelin Observatory, Svalbard during the period Nov 2015–Feb 2018. Solid and dotted lines show median and mean values, respectively, and shaded areas indicate the 25$^{\mathrm{th}}$ to 75$^{\mathrm{th}}$ percentile ranges. Data have been segregated based on the ~~updraft velocity~~ temperature at the station, ~~w~~T. The panels show data for **a** all ~~w~~T, **b** ~~$w < 1\ \mathrm{ms}^{-1}$~~$T > -4\,^{\circ}\mathrm{C}$, **c** ~~$w \geq 1\ \mathrm{ms}^{-1}$~~$T \leq -4\,^{\circ}\mathrm{C}$, and the corresponding bar charts show the number of data points per month.

low, relatively stable concentrations during the rest of the year. There are some differences compared to previous measurements at Zeppelin Observatory related to the natural variability of aerosols, for example in terms of when peak concentrations ~~occur~~

were observed (e.g., Ström et al., 2003; Tunved et al., 2013; Freud et al., 2017). Such differences could be due to annual variability, which has previously been shown to be significant (Freud et al., 2017). In addition, it should also be kept in mind that we present number concentrations exclusively during cloud events and concentrations shown are for particles above 20 nm diameter, in contrast to previous studies.

The cloud residual number concentrations, while lower than the total particle concentrations, display a similar seasonal behaviour. ~~The~~ As seen in Fig. 2a, the shape and the magnitude of the cloud residual annual cycle ~~are confirmed by ambient cloud particle measurements (cf.~~ agree nicely with the cloud particle annual cycle.

In Fig. ~~2a).~~ 8a, the overall cloud residual number concentrations range between 9 and 65 cm^{-3} (25^{th} and 75^{th} percentiles), with a median of 23 cm^{-3} (mean $\pm$ standard deviation: 51 ± 71 cm^{-3}). The corresponding total particle concentration during these cloud events is generally higher, ranging from 22 to 130 cm^{-3} (25^{th} and 75^{th} percentiles) with a median of 56 cm^{-3} (mean $\pm$ standard deviation: 107 ± 159 cm^{-3}). These numbers do not change appreciably when only clouds at temperatures above -4°C (Fig. 8b) are considered, which shows that the annual cycle of cloud residual number concentrations is driven by mostly liquid clouds.

Based on Fig. 8a, the springtime peak in cloud residual concentrations appears to lag that of the total aerosol particles by one month – the maximum cloud residual concentration occurs in May rather than in April. ~~However, this apparent shift does not appear if we only consider low updraft cases (~~That lag is not apparent in Fig. 8b ~~). The total particle concentration peak in April in Fig. 8a appears to be driven by the high total particle number concentrations in April during high updraft events (~~(warm clouds) while it does appear in Fig. 8c ~~). Most of the April data points in Fig. 8c are from the same event – a relatively thin cloud where a large difference between the cloud residual and total particle concentrations was observed. Overall, the high updraft cases are characterised by very low cloud residual number concentrations and the shape of the annual cycle is slightly different from the other two panels. This may indicate that an explanation could be the potential decrease in the GCVI sampling efficiency at high updrafts (see Sect. **??**). One should also bear in mind that the months of March and April are characterised by a low number of observations (80-100 size distribution scans), limiting the statistical significance for these months for the concurrent data.~~ (cold clouds); however, the low data coverage in March and April makes it difficult to draw any conclusions.

~~In terms of number size distribution (see Fig. 9), the cloud residual population is dominated by accumulation mode particles during most of the year. However, the Aitken mode is often also present,~~ There are only a few long-term CCN data sets from the Arctic (Jung et al., 2018; Dall'Osto et al., 2017; Schmale et al., 2018) that we can compare our measurements to, and ~~there appears to be a clear seasonality in the relative abundance of Aitken and accumulation mode cloud residuals. In January and February, the median size distributions are dominated by the Aitken mode. In spring, particularly April and May, there are very few Aitken mode cloud residualsin comparison to the number~~ they are based on a different measurement technique. Cloud residual measurements differ from standard CCN measurements in that instead of attempting to replicate in-cloud conditions inside the instrument – most notably fixed supersaturation bands in place of dynamic ambient conditions – we extract cloud particles from the air, dry them and subsequently count and size the cloud residuals. Since these other techniques cannot measure INP, we will only compare them to our liquid cloud-dominated data.

Jung et al. (2018) found that CCN concentrations correlated well with concentrations of accumulation mode ~~residuals. The size distributions then become more bimodal during summer and autumn, then in December the Aitken mode dominates again. The size distributions in Fig. 9 are normalised to highlight their shape. Non-normalised monthly cloud residual size distributions, together with concurrent total aerosol size distributions, can be found in~~ particles at Zeppelin Observatory, and that median CCN concentrations peaked in March at most supersaturation levels. This is slightly different from our measured cloud residual concentrations, which peak in April (Fig. ~~?? in the supplementary material. Note, however, that the figures show different subsets of the data – Fig. 9 shows all cloud residual data we have, while Fig. ?? is limited by the availability of concurrent data from the whole-air inlet. Generally speaking, the total aerosol size distributions and the cloud residual size distributions show similar size modes, but the total aerosol concentrationsare higher, particularly for the Aitken mode.~~

~~In Fig. 6, clear changes in the size cloud residual distributions (~~8b). However, one should keep in mind that Jung et al. (2018) considered different years (2007–2013) and did not differentiate between in- or out-of-cloud periods. In addition, Jung et al. (2018) observed for most of the year higher CCN concentrations than our cloud residual concentrations, particularly in winter, highlighting the differences between measurement techniques. Studies where particles are artificially activated, i.e. ~~shift towards smaller activation diameters) are only seen for updrafts above around 2 ms^{-1}, which is a relatively small subset of the observations. The overall shape of the monthly averaged cloud residual size distributions do not change significantly if only taking into account cases with updrafts below 1 ms^{-1} (see Fig.~~at a fixed supersaturation, are independent of the ambient meteorology and atmospheric dynamics, whereas our study inherently takes the ambient conditions into account by sampling the actual cloud droplets or ice crystals. 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 there are still some ice processes involved in the data in Fig. ~~?? in the supplementary material). As such, we will use all size distributions for all updrafts from now on.~~

8b while CCN counters only consider liquid droplet activation.

4 ~~The importance of Aitken mode particles in Arctic clouds~~

Cluster analysis

~~Figure 9 shows that Aitken mode cloud residuals, even below 30 nm in diameter, occur throughout the year at Zeppelin Observatory. In this size range, particles are often not considered to be potential CCN (nor INP), but a closer look at our measured cloud residual size distributions shows that Aitken mode particles often make up a significant part of the total cloud residual number concentration. Figure 10 shows the seasonality of the contribution of sub-100 nm, sub-50 nm and sub-25 nm particles to the overall measured cloud residual population.The sub-100 nm size range is included for illustrative purposes, since 100 nm is sometimes used as a lower size threshold for particles to be considered CCN-active for liquid clouds (Kuang et al., 2009; Yu et al., 2014; Patoulias et al., 2015). The fractions presented in~~ While Fig. ~~10 have been calculated based on daily mean cloud residual concentrations and show that particles smaller than 100 nm in diameter make up between 30 and 70 % of the total measured cloud residual number concentration in the majority of the cases. In fact, the averagecontribution (both mean and median) is close to or above 50 % in all but four months. The months where the sub-100 nm cloud residuals~~

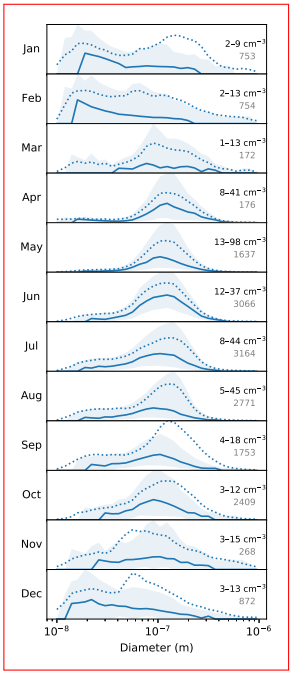

Figure 9. ~~**Monthly average cloud residual number size distributions.** Solid and dotted lines show median and mean values, respectively, and shaded areas indicate the 25^{th} to 75^{th} percentile ranges. The numbers in the upper right corner of each panel indicates the 25^{th} to 75^{th} percentile ranges of the integrated number concentrations. The grey numbers below indicate the number of data points we have per month. Concurrent cloud residual and whole-air size distributions are shown for comparison in Fig. **??** in the supplementary material.~~

~~make up a smaller fraction of the total are the months when the total aerosol particle number concentration is the highest (April through July; cf.~~ 7c indicated that the cloud residuals behave differently, on average, below $-4°$C, using a strict temperature cut introduces some problems with data availability (as seen in Fig. 8~~).~~

~~The seasonal pattern looks similar for all three cloud residual size ranges in Fig. 10. The relative contribution of Aitken~~
670 ~~mode particles to the total cloud residual number concentration increases during autumn and continues to do so until it reaches a maximum in February. Then, when the haze period starts in March and April, the contribution of Aitken mode particles begins to decrease as the number of accumulation mode particles increases. The relative contribution of sub-100 nm particles is at its lowest in April, while the sub-50 nm and sub-25 nm relative contributions continue to decrease until June. This behaviour is opposite to~~ c) and interpretation – is there really a physical reason to expect different behaviours on either side of this specific
675 temperature boundary? Cluster analysis provides a means of sorting the data based on the shape of the ~~total aerosol particles, where the summer months are the months with the highest relative contribution of Aitken mode particles due to the increased contribution of new particle formation (cf.~~ cloud residual size distributions, which allows us to study the annual cycle of behaviour without relying on an external parameter for grouping the data. For completeness, monthly average cloud residual size distributions above and below $-4°$C are shown in Fig. ~~**??**~~9.

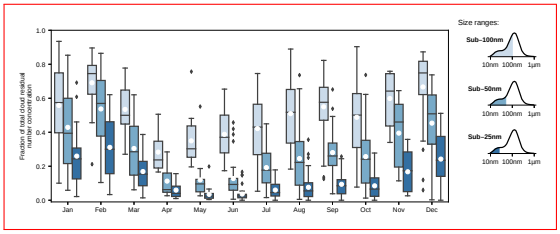

Figure 10. ~~**Contribution of small particles to the overall cloud residual population.** Box plot of daily average contributions (fraction of the total cloud residual number concentration) of Aitken mode particles to the cloud residual population. The whiskers extend no more than 1.5 times the interquartile range past the edges of the box, and data points outside that range are marked by black diamonds. Mean values are indicated by white dots. The different shades of blue indicate the cloud residual size ranges sub-100 nm, sub-50 nm and sub-25 nm diameter (see legend).~~

680 ~~To find out under which conditions the smaller residuals are present in the cloud particles, we~~ We performed k-means clustering on the cloud residual number size distributions (normalised by the corresponding total cloud residual number concentration). The results when 5 clusters are used are presented in Fig. 11. The clusters are numbered from 1 to 5 according to increasing modal diameters of the cluster average size distributions (the approximate modal diameters are 15, 30, 65, 100, and 150 nm). This order is also reflected in the ~~total number concentration~~ number concentration of both cloud residuals, cloud
685 particles, and total particles (cf. Fig. 12d–e). Cluster 2 is the most frequent cluster (27 % of the time) spread throughout the year but less in spring and early summer. This is followed by Clusters 5 and 4 (26 % and 25 %, respectively) which are more dominant in spring and summer. Cluster 3 (14 % of the time) occurs more in late summer and autumn, while Cluster 1 is the least frequent (8 % of the time) and occurs mostly during winter.

~~Two of the clusters (1 and 2 in Fig. 11) show cloud residual size distributions dominated by Aitken mode particles. The~~
690 ~~accumulation mode Clusters (3, 4, and 5)~~

Clusters 3–5

Clusters 3–5 show almost identical cloud particle number size distributions with a mode around 12 μm cloud particle diameter. The ~~Aitken mode clusters, on the other hand, are associated with larger cloud particles (Fig. 11a) and the lowest cloud particle and cloud residual number~~ cloud residual number size distributions are all dominated by the accumulation mode and have
695 a similar shape, although with different modal diameters as mentioned above. These clusters all agree quite well with the concentrations from the FM-120, with 95% of the residual concentrations being within a factor of 5 from the cloud particle concentrations (Fig. ~~12), suggesting they represent optically thin clouds with few, large droplets and/or ice crystals. This is further corroborated by the visibility distribution (Fig. 12) which shows high values, in particular for Cluster 1. Cluster 1 also stands out in that it occurs primarily during the winter months and at low temperatures (Fig. 12). No clear relationship between~~
700 ~~these two clusters and wind speed or updraft was found. The ratio between cluster mean cloud residual size distributions and corresponding mean ambient particle size distributions, for liquid clouds this would correspond to the activation ratio, are shown in Fig. 14 in the supplementary material. Cluster 1 and (to a lesser degree) Cluster 2 clearly deviate from the~~

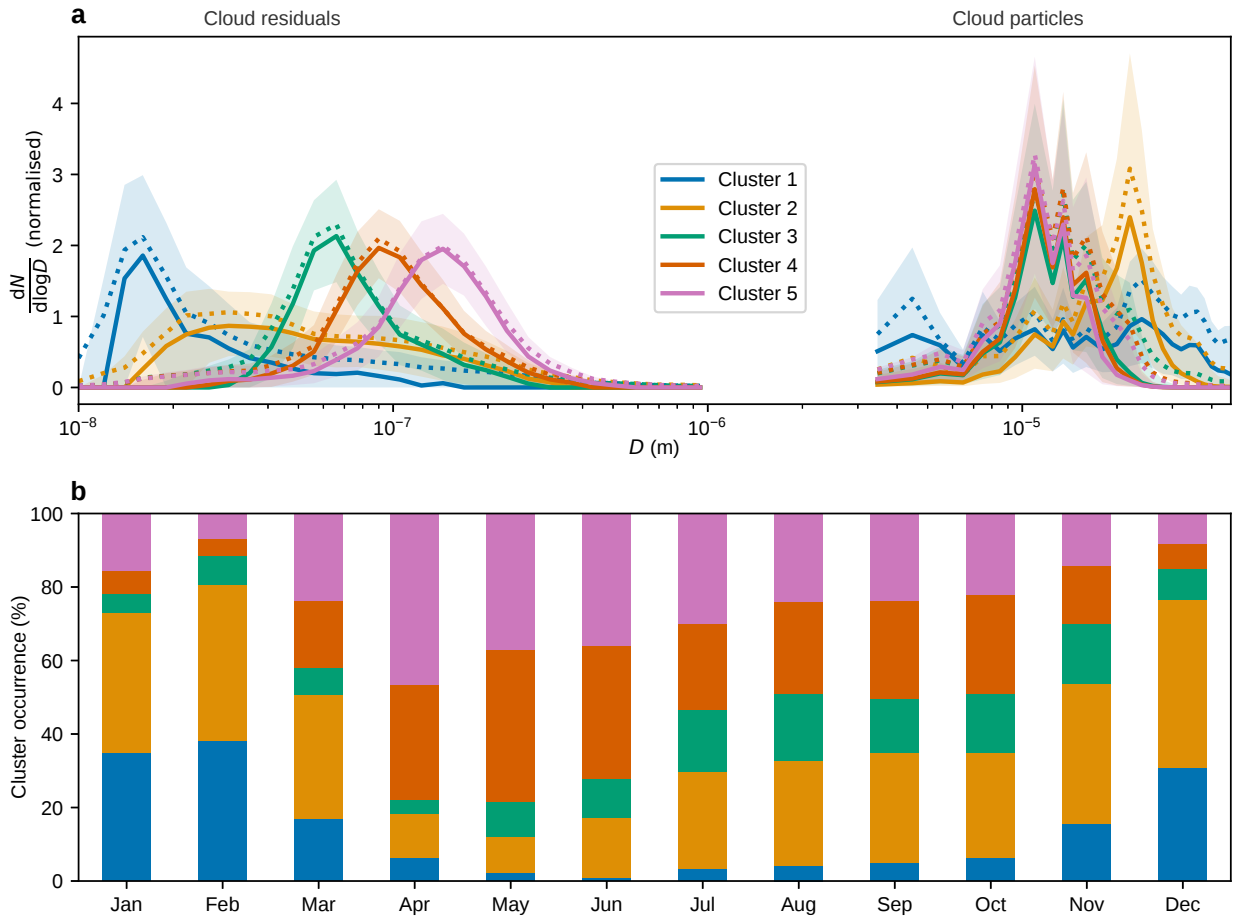

Figure 11. Results of k-means clustering of cloud residual number size distributions using 5 clusters. **a** Normalised cloud residual number size distributions for each cluster (left), and normalised number size distributions for the corresponding ~~ambient~~ cloud particle population (right). Solid and dotted lines show median and mean values, respectively, and shaded areas indicate the 25^{th} to 75^{th} percentile ranges. **b** Monthly frequency of occurrence of each cluster. ~~Additional parameters (e.g. integrated cloud residual and cloud particle number concentrations, non-normalised cloud residual size distributions, etc.) for each cluster can be found in the supplementary material.~~

~~classical Köhler theory of droplet activation assuming a size-independent chemical composition. It is possible that differences in particle composition could explain part of this behaviour (e.g., McFiggans et al., 2006; Lowe et al., 2019); however, liquid droplet activation is not the only relevant process at our site and therefore the cloud residual distributions we measure could also be related to ice processes. The fact that Cluster 1 occurs predominantly during winter and at lower temperatures than the other clusters (Fig. 12) would also be consistent with an influence from ice processes.~~

S9c–e). While all clusters occur throughout the year, these three clusters occur mainly during the warmer months, and thus correspond mainly to liquid clouds. Comparing the target classification of Cloudnet above Ny-Ålesund, at the altitude around

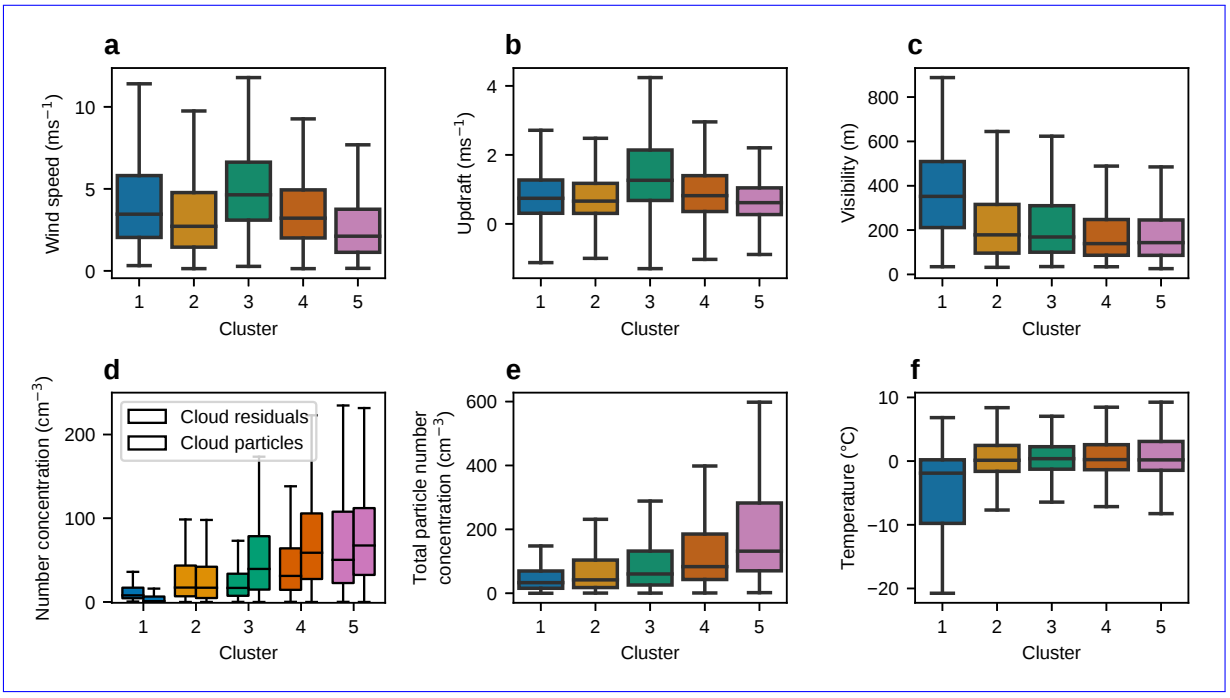

Figure 12. Additional parameters for the cluster analysis from Figure 11. The panels show the distribution per cluster of **a** wind speed, **b** updraft, **c** visibility, **d** cloud residual (solid; corrected for CVI sampling efficiency) and cloud particle (hatched; without any correction factors) number concentrations, **e** total particle number concentration and **f** temperature. The whiskers of the box plots extend to the farthest points that are within 1.5 times the interquartile range from the nearest quartile. Points that fall outside the whiskers are not shown.

Zeppelin Observatory, to the cluster analysis indeed shows a ~~higher occurrence of ice for Cluster~~ lower ratio of ice to liquid occurrence (Cloudnet category 4 divided by category 1) for these clusters compared to the other two (Fig. 13 and Fig. S10 ~~a~~ in supplementary material). Note that the Cloudnet comparison could only be made for a subset of the data (see Sect. 2.3.4 and Table S1), but the relative cluster occurrence in this subset is similar to that in the full ~~dataset. Interestingly, the ratio of ice to liquid occurrence (Cloudnet category 4 divided by category 1) decreases from Cluster 1 to Cluster 5, which is consistent with the activation ratios in Fig. 14 which appear more like classical Köhler activation (of homogeneously mixed particles) when moving from Cluster 1 to Cluster 5. However, it~~ data set. It should be noted that all Cloudnet classification categories appear in each of the clusters (Fig. S10), so the cloud residual size distribution shapes cannot be solely attributed to one cloud particle type. However, even for mixed-phase clouds the majority of the particle concentration is likely to be made up of supercooled droplets rather than ice crystals.

4 ~~Discussion~~

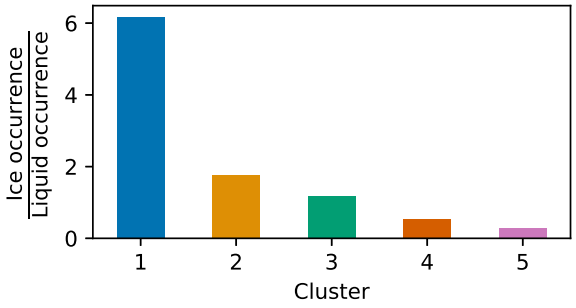

Figure 13. Ratio of ice to liquid occurrence per cluster based on Cloudnet retrieval. The bar chart shows, for each cloud residual size distribution cluster, the average ratio of pure ice to pure liquid occurrence, i.e. Cloudnet category 4 divided by Cloudnet category 1, around the altitude of Zeppelin Observatory. See Fig. S10 for a more detailed view on the average Cloudnet retrieval target classifications for each cluster.

~~Results presented in this paper are the first direct long-term measurements of size resolved cloud residual number concentrations of low-level clouds in the Arctic.It is also the first cloud residual dataset that covers more than a full annual cycle, in the Arctic and globally.It includes the important winter months, when Arctic warming is most pronounced (Maturilli and Kayser, 2017) and clouds are hypothesised to play a key role. Our measured cloud residual number concentrations generally follow the typical annual aerosol cycle previously reported for this site (Tunved et al., 2013). During the autumn and winter months, we found, in relative terms, a significant contribution of Aitken mode particles to the cloud residual number concentration~~The ratios between the cluster average cloud residual size distributions and corresponding average total particle size distributions, which for liquid clouds would correspond to the activation ratio, are shown in Fig. 14. Indeed, for clusters 3–5, the ratios are more or less S-shaped (albeit more so for cluster 5 than the other two), as would be expected from the classical Köhler theory of liquid droplet activation (assuming a size-independent chemical composition).

The modal diameters of clusters 3–5 in Fig. 11a and the $D_{50\%}$ in Fig. 14c–e correlate with the total particle number concentration (Fig. 12e) and anticorrelate with updraft velocity (Fig. 12b). A relationship between updraft velocity and $D_{50\%}$ was not clearly seen in Sect. 3.2.1; however, it could be 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. As previously stated, both an increased updraft velocity and decreased particle concentrations can allow smaller particles to activate (assuming the meteorological conditions are otherwise the same).

Clusters 4 and 5 together make up just over half of all data, and they are also associated with the highest total particle number concentrations (cf. Fig. 12d–e). Thus, these clusters have a large influence on the overall annual cycle of particle number concentrations. From Fig. 11b, it can be seen that the occurrence of cluster 5 peaks in April, when total particle concentrations peak (cf. Fig. 8). This indicates that perhaps cluster 5 represents a typical cloud residual size distribution associated with the Arctic Haze.

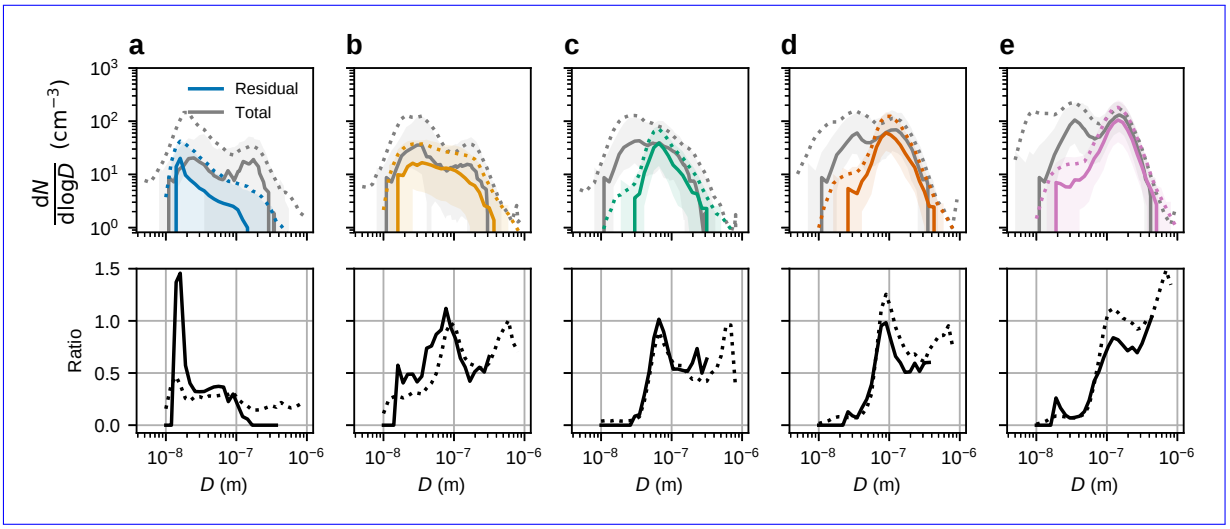

Figure 14. Size distributions and activation ratios for the clusters from Figure 11. The top row shows cloud residual size distributions (in colour) and the corresponding total particle size distributions (grey). Solid and dotted lines show median and mean values, respectively, and shaded areas indicate the 25th to 75th percentile ranges. The bottom row shows activation ratios calculated from the median (solid) and mean (dotted) distributions. Columns **a–e** show Clusters 1–5, respectively.

~~Cloud residual measurements differ from standard CCN measurements in that instead of attempting to replicate in-cloud conditions inside the instrument – most notably fixed supersaturation bands in place of dynamic ambient conditions – we extract cloud particles from the air, dry them and subsequently count and size~~

Clusters 1 and 2

Clusters 1 and 2 clearly deviate from the other three clusters, both in terms of the ~~cloud residuals. While there are only a few long-term datasets from the Arctic (Jung et al., 2018; Dall'Osto et al., 2017; Schmale et al., 2018) that we can compare to, this difference in measurement techniques seems to be important. Jung et al. (2018) found that CCN concentrations correlated well with concentrations of accumulation mode particlesat Zeppelin Observatory,~~ shape of the cloud residual size distribution and the cloud particle size distribution. They are also very different from each other, but what they have in common is that they show cloud residual size distributions dominated by Aitken mode particles. While cluster 1 consists almost entirely of Aitken mode particles, cluster 2 is bimodal with two broad size modes (i.e. it contains both Aitken and accumulation mode particles). Cluster 1 is not related to any clear cloud particle size mode but, in relative terms, it is associated with larger cloud particles than clusters 3–5 (Fig. 11a). Cluster 2 is also associated with larger cloud particles with a mode around 23 μm (cf. 12 μm for clusters 3–5). Clusters 1 and ~~that median CCN concentrations peaked in March at most supersaturation levels. This is different from our measured cloud residual concentrations, which peak in May. However, one should keep in mind that Jung et al. (2018) considered different years (2007–2013)and did not differentiate between in- or out-of-cloud periods. In addition, Jung et al. (2018) observed for most of the year higher CCN concentrations than our cloud residual concentrations,~~

particularly in winter, highlighting the differences between measurement techniques. Studies where particles are artificially activated, i.e2 also have the lowest cloud particle and cloud residual number concentrations of the clusters (Fig. at a fixed supersaturation, are independent of the ambient meteorology and atmospheric dynamics, whereas our study inherently takes the ambient conditions into account by sampling the actual ambient cloud droplets 12d). These factors suggest they represent thin clouds with few, large droplets and/or ice crystals. 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 ice processes are involved while CCN counters only consider liquid droplet activation.This is further corroborated by the visibility distribution (Fig. 12c) which shows high values, in particular for cluster 1. No clear relationship between these two clusters and wind speed or updraft was found (Fig. 12a–b).

The importance of Aitken mode particles for Arctic clouds has previously been shown (e.g., Leaitch et al., 2016; Koike et al., 2019; Korh however, by indirect means or model studies.Our results support these findings with direct measurements of cloud residuals. Furthermore, we find that Aitken mode particles also play an important role in wintertime clouds at Zeppelin Observatory, while previous observations have focused on the Arctic summer months. The clear seasonality we observe in the relative contribution of small particles to Cluster 1 is the one that stands out the most. The shape of the cloud residual number concentration could partly be explained by the interplay between aerosol particle sources, sinks, meteorology and condensible water vapour. In late autumn and winter , aerosol particle concentrations decrease rapidly size distribution is peculiar, with peak concentrations occurring below 20 nm particle diameter. Cluster 1 occurs primarily during the winter months and at low temperatures, and at considerably higher visibility and lower cloud particle concentrations than the other clusters (Fig. 8)and the Arctic atmosphere becomes drier (Maturilli and Kayser, 2017). However, if the decrease in condensation sink, due to reduced particle concentration, is larger than the decrease in water vapour, there will be, in relative terms, more water vapour available for fewer particlesin winter. These conditions allow for higher supersaturation to be reached and smaller particles to be activated (assuming no strong seasonal cycle in the updraft velocity as observed here, see 12). These conditions would be consistent with ice or mixed-phase clouds in the winter. The Cloudnet analysis shows that cluster 1 has by far the highest occurrence of ice crystals compared to liquid droplets (Figs. 13 and S10a). Interestingly, the ice to liquid ratio decreases from cluster 1 to cluster 5, which is consistent with the activation ratios in Fig. 14 which appear more like classical Köhler activation (of homogeneously mixed particles) when moving from cluster 1 to cluster 5.

The shape of the cloud residual size distribution of cluster 1 compared to the ambient particle size distribution (cf. Fig. S1) . In other words, the winter season at Zeppelin Observatory falls into the CCN-limited cloud–aerosol regime that has previously been reported for the summertime High Arctic (Mauritsen et al., 2011; Leaitch et al., 2016). However, this only applies to liquid clouds, while the cloud residuals we measure could correspond to either CCN or INP. Unfortunately, no cloud phase data are available for our measurement period but, by proxy of the Cloudnet target classification from above Ny-Ålesund, we have shown that the cloud residual size distributions dominated by the very smallest particles are likely to be influenced by ice processes.

Some 14a) reveals that the accumulation mode particles do not activate. In mixed-phase clouds, supercooled droplets outnumber ice crystals, often by orders of magnitude (e.g., Young et al., 2016), so we should be seeing accumulation mode

cloud residuals stemming from the supercooled droplets in addition to the small particles. Verheggen et al. (2007) observed a decreased activated fraction (of particles larger than 100 nm) with decreasing temperature similar to our observations (see Fig. S11), which they attributed to the Wegener-Bergeron-Findeisen process, i.e. evaporation of liquid droplets promoting ice crystal growth. This might be an explanation for the missing accumulation mode in cluster 1. The cold temperatures and very low cloud particle concentrations associated with cluster 1 may also be consistent with pure ice clouds. However, some of the cloud residuals we have measured, in particular those in ~~Clusters~~ cluster 1~~and 2~~, are much smaller than typical INP (Hoose and Möhler, 2012; DeMott et al., 2010) (or indeed CCN). ~~Yet residual~~ Residual size distributions with a ~~similar shape~~ very similar shape as the one of cluster 1 have 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). Although those studies used different techniques and sampled different cloud types, it could be an indication that cluster 1 is the result of ice particles sampled by the GCVI.

Nevertheless, the question arises whether these small particles are really cloud residuals, or if they are measurement artefacts. As discussed in Sect. 3.1.3, artefacts in the form of ice crystal shattering cannot be fully ruled out. Large crystals are expected to be more prone to shattering, and indeed clusters 1 and ~~droplet residualsdown to 25~~2 are related to larger cloud particles than the other clusters (Fig. ~~nm diameter have previously been observed by GCVI measurements (Schwarzenboeck et al., 2000) and predicted in model studies (Gérémy et al., 2000; Korhonen et al., 2008)~~11a). Cloudnet does not distinguish between cloud ice and precipitating ice, so we could also be dealing with snowflakes. The average cloud particle size distribution associated with cluster 1 is rather flat with no obvious mode (Fig. 11a), and it is also associated with very low cloud particle concentrations (Fig. 12d). This could just be noisy measurements in the fog monitor during snowfall, and would indicate that cluster 1 is influenced by snow. Cluster 2, on the other hand, has a clear cloud particle size mode, although at a larger diameter compared to cluster 3–5 (Fig. 11a), and the cloud residuals are therefore much less likely to stem solely from precipitation.

~~A question that arises is where these small particles come from .~~ The median cloud residual concentration is slightly larger than the median total particle concentration for cluster 1 around 20 nm (Fig. 14a), which would suggest that there is a risk of crystal shattering artefacts. As shown above, Figure 2b revealed two groups of data, where one showed a discrepancy between measured cloud residual and cloud particle concentrations as would be expected with this type of artefact. The same figure separated by cluster (Fig. S9) shows that this group of data is overrepresented in clusters 1 and 2, which speaks in favour of the crystal shattering hypothesis as well. 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 S12a; note, results do not change if we are even stricter, i.e. within 1:2 and 2:1). Also note that a cloud particle mode then appears for the small particle cluster (Fig S12a), indicating a decreased relative influence from snow. This 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.

The presence of 20 nm particles is also observed in the whole-air inlet for the same times as cluster 1 occurs (Fig. 14a). The whole-air inlet is very different from the GCVI inlet (e.g. in terms of flows, velocities and how particles move inside the inlets), so it is unlikely that both inlets would produce artefacts with the same frequency and particle size. Yet, if the small

particles are real, where do they come from? In the Arctic and marine boundary layer, the presence of particles below ∼50 nm is most often associated with new particle formation (Ström et al., 2003; Tunved et al., 2013) or even primary emissions of sea spray particles (Ovadnevaite et al., 2011). However, these sources are unlikely to explain the presence of small particles during winter, when there is reduced or no sunlight (i.e. no photochemistry), less biological production and most of the sea surface is covered by ice (Dall'Osto et al., 2017; Sharma et al., 2012). Other potential sources can be long-range transport, but the lifetime of Aitken mode aerosol particles in the boundary layer is rather limited, or entrainment from the free troposphere. However, this is purely speculative and future studies are needed to investigate the exact sources and chemical nature of these small particles.

~~As the remote-sensing results suggest, the ice phase appears at the height of Zeppelin Observatory predominantly during mixed-phase cloud conditions~~At this point, it is important to point out that, even barring artefacts, a cloud residual does not necessarily respond directly to a CCN or INP. Cloud residuals can also be nuclei that have undergone processing inside the cloud (be that chemical or physical), and can contain material from e.g. riming or aerosol particles that have been scavenged by the cloud particles. Unfortunately, we have no way of distinguishing between these particle types, especially since the FM-120 cannot differentiate between cloud droplets and ice crystals. It could potentially be that the crystals we measure are the result of secondary ice formation processes (Field et al., 2016), which has been suggested in a model study to be important for Arctic stratocumulus clouds (Sotiropoulou et al., 2020). ~~This could include a distribution of the original CCN or INP material to the ice splinters, which act as new nuclei to further ice particle formation.~~ In other words, the cloud residuals we measure ~~do not have to correspond to single CCN or INP but may also~~ may be fragments of ~~these~~ nuclei and/or scavenged particles, which would explain their small size. ~~The shape of the cloud residual size distribution of Cluster~~

Since secondary ice formation happens before the cloud particles enter our inlets, these particles should also be seen by the fog monitor. This is often not the case for cluster 1~~compared to the ambient particle size distribution (cf. Fig.~~, as seen in Fig. ~~14a) reveals that the accumulation mode particles do not activate. This points towards a possible water vapour transfer to the ice splinters via the Bergeron–Findeisen process causing a larger concentration of interstitial aerosol particles.However, it should be noted~~ S9, which means that secondary ice particles cannot be the only reason for the small residuals we observe ~~, as it would not explain why the cloud residual and cloud particle concentrations do not always agree during these cases (see e.g. Fig.7a)~~ (unless the ice crystals are undersampled by the fog monitor, see Sect. 3.1). ~~There are other processes, such as riming, which could be consistent with both small residuals and a discrepancy between cloud residual and cloud particle concentrations.Additionally, since mixed-phase clouds are concerned, part of the cloud residuals will of course also come from liquid cloud droplets. It~~ Cloud observations at mountain-top stations such as Zeppelin Observatory may also be influenced by surface processes (e.g. blowing snow) that could increase the ice crystal concentrations (Beck et al., 2018), but this, too, should be seen by the fog monitor and as discussed earlier, no clear dependency on wind-speed has been observed. As stated earlier, it is not possible to ~~tell if a given cloud residual is a result of liquid droplet activation (CCN), ice nucleation (INP), or secondary processes (CCN/INP fragment)~~ translate the cloud residual data to CCN, INP, etc without further detailed information on cloud phase, structure and origin. The cloud phase is an important parameter and should as such be added in future studies.

~~Although an overall good agreement between ambient cloud particle and cloud residual number concentrations is found, one has to keep in mind that measurement artefacts can still not be fully excluded given the complexity of our observations.The initial data set was carefully screened for malfunctioning of instrumentation and local contamination.A thorough assessment of potential artefacts and instrument uncertainties was made in Sect~~In summary, it seems likely that cluster 1 is significantly influenced by snow and ice. It is difficult to say to what extent the signal is caused by crystal shattering artefacts as compared to other processes, but cold temperature outliers (cf. Sect. 3.1.3) make up roughly 45% of cluster 1 (cf. ~~3.1.While there were some cases where the agreement between the GCVI and the FM-120 was clearly worse, we did not discard these data because we were unable to prove that the disagreement was only caused by artefacts in the GCVI. In addition, the disagreement is not completely random~~ Fig. S9a). These data should be treated with caution, but there are some plausible physical explanations for the presence of small particles when the agreement with the fog monitor is better, e.g. secondary ice processes, yet further measurements would be needed to verify this. The possibility that such processes would show a signal similar to shattering artefacts is an important consideration when analysing GCVI data from ice or mixed-phase cloud conditions. However, from an aerosol activation perspective, it is irrelevant whether the snow or ice crystals shatter before or after they enter the inlet – ~~data points at cold temperatures are overrepresented (see Figs~~in both cases, the resulting cloud residuals do not represent cloud nuclei.

Cluster 2 is also Aitken mode dominated, and occurs throughout the year (27 % of the time, or 13 % of the time if we only consider $T > 0$°C). Unlike cluster 1, cluster 2 is much less likely to be affected by snow artefacts. While the exact contribution is difficult to quantify, cold temperature outliers only make up about 13% of cluster 2 (cf. ~~2 and S5).Thus, removing these points would have introduced a (potentially unjustified)bias into our analysis.Despite the uncertainties, we believe that our results are reliable enough to show that small particles are likely contributing to the formation of mixed-phased clouds at Zeppelin Observatory. This is especially important during the dark period when overall aerosol concentrations are low and even small changes in available CCN and INP will have a strong impact on cloud properties.~~

~~Our study presents a unique seasonal picture of aerosol particle activation in clouds in a polar environment and is the first long-term study of~~ Fig. S9b), i.e. there is a significantly better agreement with the fog monitor than for cluster 1. Cluster 2 is also different from clusters 3–5 but, in contrast to cluster 1, it was observed more homogeneously throughout the year. In further contrast to cluster 1, the meteorological parameters related to cluster 2 are not distinctly different from those related to clusters 3–5 (cf. Fig. 12). This means that cluster 2 was also observed during sampling conditions when we can safely rule out the influence of mixed-phase clouds and ice crystals. Many of the caveats listed above related to cluster 1 thus do not apply to cluster 2 to the same extent. In addition, cluster 2 does not show the lack of accumulation mode particle activation that complicated the interpretation of cluster 1. Hence, the Aitken mode cloud residuals in cluster 2 very likely contain activated aerosol particles. Similar findings were reported in previous CVI measurements (Schwarzenboeck et al., 2000), although not in the Arctic. ~~Activation of aerosol particles in~~ In the Arctic, activation of Aitken mode aerosol particles has been shown by indirect means and model studies (e.g., Leaitch et al., 2016; Koike et al., 2019; Korhonen et al., 2008).

4 Conclusions

Results presented in this paper are the first direct long-term measurements of size-resolved cloud residual number concentrations of Arctic low-level ~~clouds in the Arctic is strongly coupled to the annual variability of aerosol particle number and size distribution, meteorology and the availability of water vapour. We have demonstrated that the use of CCN proxies with fixed size limits (e.g. 100 nm diameter)or accumulation mode aerosol particles is incorrect for the Arctic environment, where smaller particles act as CCN, supporting the results of previous studies. For large parts of~~ clouds. It is also the first cloud residual data set that covers more than a full annual cycle, in the Arctic and globally.

We conducted a thorough evaluation of the GCVI measurements by comparing them to cloud particle size distributions as measured by an FM-120 fog monitor, as well as to total particle size distributions measured behind a whole-air inlet. We derived a correction factor for the cloud residual measurements based on the cloud particle data and the experimentally determined sampling efficiency of the GCVI. For warm clouds, we could also derive a correction factor by comparing cloud residual and whole-air accumulation mode particle concentrations (under the assumption of liquid droplet activation with no size-dependent chemical composition), and we found that both methods agreed remarkably well (within one standard deviation). Our data set includes the winter months, when Arctic warming is most pronounced and clouds are hypothesised to play a key role. However, as it turns out, the ~~year, and especially during the dark period~~winter months are not entirely straight-forward to analyse. We identified a group of data at cold temperatures where the cloud residual and cloud particle concentrations did not agree well. It is likely that this is a result of snow or ice crystal shattering artefacts. However, these points are a small percentage ($\sim$7–8%) of the total data and the majority of the data are not affected by potential sampling artefacts.

Our measured cloud residual number concentrations generally follow the typical annual aerosol cycle previously reported for this site. For pure liquid clouds (T$>$ 0°C), we observed ~~a large relative contribution of Aitken mode particles to the~~ activation diameters ($D_{50\%}$) in the range of 58–78 nm. The cluster analysis of cloud residual size distributions showed a $D_{50\%}$ dependence on updraft for liquid clouds (clusters 3–5). There was also a clear correlation with the total number concentration of aerosol particles. Although we cannot clearly disentangle the influence of these parameters, our analysis indicates that it is perhaps more likely that the number concentration drives the difference in $D_{50\%}$. A clear relationship between a decreasing total particle number concentration and a decrease in $D_{50\%}$ was also observed when the cloud ~~residuals, which could be explained either by a strong CCN-limited regime or ice processes, possibly including secondary ice formation. In winter,~~ residual size distributions were binned by temperature.

In April–October, 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. In November–March, we found, in relative terms, a significant contribution of Aitken mode particles to the ~~Arctic exhibits the strongest warming trend and clouds are believed to play an important role in this process. Even subtle changes in aerosol particle number concentrations during the dark period in~~ cloud residual number concentration. However, the presence of the ice phase and snow complicates matters. The mode of smallest particles we observed (cluster 1) is most likely due to artefacts of crystal shattering within the wind tunnel of the ~~Arctic can result in large effects on cloud microphyscial properties and thus also perturb cloud-related warming effects by~~

~~changing the radiation balance in the infrared spectrum. The climatology presented here provides a new benchmark dataset for further model–measurement evaluation exercises to improve the representation of low-level clouds in Earth system models. Our work also shows the importance of focusing more research in the Arctic on the dark period. We have demonstrated the experimental complexity involved in aerosol-cloud interaction research, highlighting the strengths and weaknesses of sampling~~ GCVI or caused by fragments of cloud nuclei or scavenged particles created through secondary ice multiplication processes. With our instrumental set up, the contribution of these different processes cannot be confidently quantified. As far as cluster 2 is concerned, while artefacts and ice processes cannot be completely ruled out, we believe that the majority of the signal is real and shows new experimental evidence of the activation of aerosol particles down to ~ 20 nm in the Arctic, confirming results from previous experimental and modelling studies.

In-situ sampling of cloud droplets and ~~crystals by means of the GCVI technique. The direct measurementsof cloud residuals provide a valuable new perspective on Arctic CCN and INP , but information about the cloud particle phase and~~ ice crystals are a complex challenge. Detailed cloud phase measurements, i.e. the ratio of ice crystals to liquid droplets within the cloud and close to the GCVI, using a more sophisticated cloud probe will be needed to better understand the relative importance of CCN / INP and the importance of other related in-cloud processes. 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. In addition, detailed and size-resolved chemical composition measurements of the sampled cloud residuals and the contribution of supermicron particles would help to better understand the ~~residual chemical composition would be necessary to be able to disentangle and better understand all the processes and particle sources involved in~~ sources and processes related to low-level Arctic cloud formation.

Data availability. The data of this study will be available on the Bolin Centre Database (DOI and link will be added later). The Cloudnet data are available on the Cloudnet website (http://devcloudnet.fmi.fi/)

Author contributions. LK analysed the data and wrote the manuscript. PZ and RK designed the study, carried out the observations and contributed to writing the manuscript. MK provided the cloud droplet distribution data. KE provided Cloudnet data. All authors discussed the results, read and commented on the manuscript.

Competing interests. PZ and RK are currently acting as co-editors in ACP. The authors declare no further competing interests.

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