Aerosol and dynamical contributions to cloud droplet formation in Arctic low-level clouds
Ghislain Motos et al.

Reply to RC1: 'Comment on egusphere-2023-530', Anonymous Referee #1, 05 Jun 2023.

We would like to thank Anonymous Referee #1 for their constructive comments and thorough efforts in helping to improve the quality of the paper. We took consideration of their comments and remarks and modified the manuscript as detailed below.

Comment:
My main criticism is that the authors rely heavily on just 4 days of tethered balloon data to conclude that cloud liquid water content is not significantly involved in the glaciation process. It is unclear how representative those balloon data are of conditions at the site or of the Arctic in general. However, given the low sample number, I suggest the authors make it very clear that while this hypothesis is supported by these limited data, further study is needed to verify this hypothesis.

Reply:
The insensitivity of droplet number to the glaciation fraction for only the clouds sampled was already noted in the original text. We emphasize this further in the revised text. Line 32 is modified as follows:
We evaluate the parameterization and the droplet numbers calculated through a droplet closure with in-cloud situ measurements taken during 9 flights over 4 days. A remarkable finding is that, for the clouds sampled in situ, closure is successful in mixed-phase cloud conditions regardless of the cloud glaciation fraction. This suggests that ice production through ice-ice collisions or droplet-shattering may have explained the high ice fraction, as opposed to rime-splintering that would have significantly reduced the cloud droplet number below levels predicted by warm cloud activation theory.
The following sentence was modified in the conclusion, at line 448:
“Although the measurements were taken over 4 days only and may not be representative of the whole year, they suggest that riming was not taking place in any significant amount, leaving room for ice-ice collisions and droplet shattering (alongside with WBF) as the main mechanisms of glaciation, in addition to primary ice production.”

Comment:
L24: “The relationship between updraft velocity and the limiting cloud droplet number agrees with previous observations of various types of clouds worldwide, which tends to confirm the universality of this relationship.” I suggest rewording to “… which supports the universality…”

Reply:
The suggested modification was made (this is actually at Line 46 and not 24).

Comment:
L73: Please consider noting that the Mioche et al., 2015 values are historical - as the Arctic continues to warm, these cloud phase statistics might become increasingly outdated.

Reply:
We consider that 2015 data are not “historical”, even though the climate of the region is changing rapidly. These statistics are thus very likely still relevant today. We included the following text:
“Although these statistics can quickly shift owing to the fast warming of the Arctic, they reflect the recent past and give a good indication of the current conditions.”

Comment:
L79: As I understand it, Zeppelin is also subject to air masses from N. America blowing up periodically through the Fram Strait.

Reply:
We included the following addition to the text:
“Differences to other Arctic stations can be partly related to Zeppelin being located close to the European and American continents, in a sector influenced by warm oceanic currents (Gulf stream) and warm air intrusions, in contrast to other stations surrounded by ice-free ocean. Zeppelin is also often located in the free troposphere (FT; Ström et al., 2009).”
Comment:
L162: Please define “non-refractory” for readers unfamiliar with the ToF-ACSM.
Reply:
The following definition was included:
“The mass concentration of non-refractory bulk aerosol (i.e., species that evaporate rapidly at a temperature of 600°C under vacuum conditions) was measured by a time-of-flight aerosol chemical speciation monitor”.

Comment:
Section 2.2.4: Please provide more information on how holographic image data were sampled on the tethered balloon. When and how often were the data taken? Up to what height? What is the uncertainty in the measurements? Are the data of equal quality in all conditions?
Reply:
Detailed information about the data taken by the holographic image on the tethered balloon can be found in Pasquier et al. (2022a). The microphysical properties of clouds were measured by the tethered balloon system for five days in November 2019 and one day in April 2020 at a height of up to 1000 m above ground. The measurement platform including the holographic imager was hanging 12 m below the tethered balloon. The classification of cloud droplets and ice crystals is performed based on their shape, using a convolutional neural network trained and fine-tuned on cloud particles from holographic imager. All ice crystals were manually classified into habits. The smallest detectable ice crystals are 25 µm, and all particles below this threshold are classified as cloud droplets. The uncertainty in the concentration of ice particles can be estimated with ±15% for ice crystals smaller than 100 µm and ±5% for ice crystals larger than about 100 µm (Beck et al., 2017). For cloud droplets, the uncertainty is estimated to be ±6%, as determined for the classification with the convolutional neural network in Touloupas et al., (2020). All ice crystals were manually classified into habits.

The corresponding paragraph was modified to the following:
“Cloud particle concentrations were sampled with the Holographic cloud Imager for Microscopic Objects (HOLIMO; Beck et al., 2017; Ramelli et al., 2020) at a height of up to 1000 m above ground for five days in November 2019 and one day in April 2020. HOLIMO can image an ensemble of cloud droplets (with diameter above 6 µm) in a three-dimensional sample volume of about 15 cm³. A convolutional neural network trained and fine-tuned on cloud particles from holographic imagers is used to identify the cloud droplets from artifacts and ice crystals (Touloupas et al., 2020) based on their shape. The smallest detectable ice crystals are 25 µm, and all particles below this threshold are classified as cloud droplets. The holographic imager was attached below the tethered balloon system HoloBalloon (Ramelli et al., 2020; Pasquier et al., 2022). Detailed information about the data taken by the holographic image on the tethered balloon can be found in (Pasquier et al., 2022a).”

Comment:
L214: “Chloride and sodium are assumed to be the only compounds predominantly present in the coarse mode…”
What about mineral dust? There are thought to be sources of local dust nearby (e.g., Fig. 5, Tobo et al. 2019).
Reply:
The text was changed to:
“Among the species measured by the ACSM, chloride and sodium are assumed to be the only ones predominantly present in the coarse mode. However, the two DMPSs […]”

Comment:
L251: “…we decided to discard uSonic data when the wind direction was between 335 and 15 degrees, so that any droplet calculation made is more representative of regional conditions than specific conditions at Zeppelin during strong orographically driven updrafts.” Good that you made this comparison, and removed those data from that analysis for this particular question - this makes the study stronger. For comparison, it would be helpful to add a related panel in the same supplemental figure that shows the relationship between the wind lidar and uSonic when those data were removed.
As a thought for future work, comparing your data with MOSAiC data (taken during the same time frame at a different Arctic location) might give extra context for your conclusions.
Reply:
A second panel was added to Figure S1 in which data between 335 and 15 degrees are removed. The new legend reads:
“Figure S1. Comparison of updraft velocity measured over the whole campaign by the wind LiDAR and the uSonic. Panel a) includes all data between October 1st, 2019 and May 1, 2020. The same data are shown in Panel
b) but data points corresponding to wind directions between 335 and 15 degrees were removed.”. Thank you for the future work suggestion as well.

Comment:
L269: “Although with a shift of a month for the high concentration plateau, these measurements are in good agreement with the annual cycle of particle concentration reported by Tunved et al. (2013) averaged over ten years from 2000 to 2010 at Zeppelin.” Please state what size ranges Tunved et al reported, and how that compares to the observations reported in Fig. 1.

Reply:
The corresponding statement was modified to the following:
“These measurements are in good agreement with the annual cycle of integrated particle concentration at Zeppelin reported by Tunved et al. (2013), who measured particle number size distribution between 20 and 630 nm before 2005 and between 10 and 790 nm after, and averaged the results over ten years from 2000 to 2010. However, we note a one-month lag in the appearance of the high concentration plateau (reported from April to July, whereas it appears from the beginning of May to the end of August in the present study).”

Comment:
Sections 4.2 and Fig. 2: The Dact (minimum diameter required for an aerosol particle to activate to a cloud droplet) estimates are really valuable information. However, the hygroscopicity portion of the work is a weak point because kappa was not directly measured, there were a lot of assumptions in its calculation, there was not great agreement between the two methods where kappa was estimated (Fig. S3b), and because there is a lot of variability in kappa values across other sites in the Arctic (section 4.2). It would be helpful to mention upfront here that this issue is partially addressed via a sensitivity analysis in section 4.3.

Reply:
We agree that it would have been a good quality control to compare filterpack-derived with CCNC-derived hygroscopicity data. This was unfortunately not possible. However, the agreement between filterpack-derived and ACSM-derived hygroscopicity is relatively good, since almost all data points agree within 50% - and its uncertainty does not translate to significant uncertainty in predicted droplet number (as shown by Fig. S5). We however consider that the weak point of our study is the fact that we don’t have any measurement of sodium and chloride, which we estimate to be mostly in the coarse mode. This is the reason why we performed a sensitivity analysis on that point. These points are already largely detailed in the last paragraph of Section 4.3, so we did not make any additional change to the manuscript.

Comment:
L305: “During periods of rain (noted in Fig. 1a), the aerosol load was strongly reduced, directly implying sharp decreases in Nd and Dact which impact the other parameters.” There is so much data shown (a good thing!) that the differences between periods of rain vs. other times is lost. It would be helpful for the reader if the authors were to add an extra figure (e.g., a box plot) showing these differences more clearly.

Reply:
We consider that the effect of wet scavenging on aerosol and droplet concentration has been well studied in the literature, and that this result is largely secondary in the context of the present study. Adding a dedicated figure could distract from the most relevant and novel points. However, we agree that the 3 panels of Figure 1 have too much data, which makes it hard to interpret any trend. We thus replaced the 3 panels by 3 seasonal histograms in Figure 2 and moved the initial plots to the Supplement (Figure S4).

Comment:
Section 4.3: The authors see evidence of high levels of supersaturation, low aerosol concentrations, and small particles that activate in fall and early winter, but not later when there are more polluted particles in the atmosphere. As context for this finding, I wonder if it would be worth commenting on how representative of the larger Arctic such conditions in Svalbard fall/early winter would be and what this finding might mean for the Arctic at large? Based on aerosol observations (e.g., Schmale et al., 2022) and model data (e.g., Eckardt et al., 2015), my guess is that many locations/times would not meet the low aerosol conditions criteria required and found in Svalbard during the fall/early winter time period. The high supersaturation requirements might reduce the locations where this process could matter even more, especially since one might expect updraft velocities over
sea ice to be smaller than over the open ocean near Svalbard. I'd be interested to know the authors thoughts on this.

Reply:
As stated in the introduction, measurements of particle size distribution performed at different Arctic sites such as Barrow in Alaska, Tiksi in Siberia, Nord and Alert in Northern Greenland showed similar patterns. (e.g., Croft et al., 2016; Freud et al., 2017). However, it is true that the fall minimum in particle number concentration might be slightly lower at Svalbard due to the presence of open ocean and warm air masses which enhance precipitation and wet scavenging. This is confirmed by a slightly lower aerosol optical depth (AOD) over Spitzbergen than over the northern coast of Canada or Siberia, as shown by Xian et al. (2022). However, this minimum is not very pronounced and not untypical of the low Arctic, and thus not specific of Zeppelin. Global model simulations indicate that maximum cloud supersaturation is mainly dependent on latitude (Pringle et al., 2009; Matsui and Liu, 2021), with similar values at latitudes above 70 degrees North (between about 0.3 to 0.7, in agreement with our results). These results speak in favour of representativeness of cloud formation regimes over the Arctic region.

The difference of updraft velocity between sea ice versus open ocean is a good point, and past/upcoming missions that will provide such data (e.g., MOSAIC, ArtOfMelt) will help constrain the distributions of aerosol and vertical velocity required to provide such an assessment. That said, the expectation is that droplet formation in such regions, owing to the higher aerosol and lower vertical velocity, would be in the lower supersaturation regime. This is also noted in the revised manuscript in the Conclusion section. We agree that the vertical updraft velocity over ice might be higher than over water, mainly because of the large temperature difference between the ocean versus the land in fall / early winter. This causes the planetary boundary to be shallower and more stratified over ice than water. For Spitsbergen specifically, the mountains may trigger gravity waves or orographic up- or downflows. However, over some weeks, all these additional vertical movements may cancel out. Hence, we do not expect a large net effect on the vertical winds caused by the differences in vertical updraft velocity. No change was made.

Comment:
L360: “Note that the three values of $\sigma_w$ chosen here are representative of the stratiform cloud conditions typically prevailing in the Arctic.” Reference?
Reply:
We included the following reference: Shupe et al., Vertical Motions in Arctic Mixed-Phase Stratiform Clouds, Journal of the Atmospheric Sciences, 2008.

Comment:
L376: “extremely low” Please be more quantitative here. Do you mean < 0.1%?
Reply:
We mention extremely low updraft velocity here, so we mean below 0.1 m/s. In our opinion, the beginning of the sentence is clear enough on that point. To make it even clearer, we modified the text to the following: “In addition, it is worth noting that when applying the same analysis with an assumption on $\sigma_w$ larger than 0.1 m s$^{-1}$ (i.e., 0.2 or 0.3 m s$^{-1}$; middle and right panels in Fig. 3) and even higher (not shown), the 0.1% $S_{\text{max}}$ threshold is not reached at all, neither in winter nor in spring, indicating that vertical velocity-limited conditions cannot be found if the turbulence of the boundary layer is not extremely low.”

Comment:
L416: Please clarify. E.g., “(ice water content as high as 90%)”, if that is what was meant here.
Reply:
We modified the text to the following: “Nevertheless, Fig. 5 provides evidence that even for a degree of glaciation (i.e., the fraction of cloud water that is in the form of ice) as high as 90%, cloud parcel activation theory can predict $N_d$ to within 50% of observations.”

Comment:
L416–422: This finding is very interesting, but the authors are overstating its significance. To agree with this conclusion, readers would need to have confidence that many clouds were measured from the data in Fig. 5, and these clouds were representative. As best I can tell, this finding is based on just 4 days of tethered balloon data.
Reply:
The four days of measurement are mentioned in this paragraph. We modified this paragraph to the following:

“Mixed-phase clouds tend to rapidly glaciate (i.e., convert to pure ice clouds) due to the Wegener-Bergeron-Findeisen (WBF) process or riming (Korolev et al., 2017), which can effectively transfer mass from the liquid to the ice phase. Nevertheless, Fig. 5 provides evidence that even for a degree of glaciation (i.e., the fraction of cloud water that is in the form of ice) as high as 90%, cloud parcel activation theory can predict $N_d$ to within 50% of observations. This implies that significant amounts of glaciation over the duration of the HoloBalloon flights, and possibly over spring and fall may have occurred through processes that do not deplete droplet number, e.g. WBF that is promoted by secondary ice production (SIP) through ice-ice collisions or droplet-shattering (Field et al., 2017; Korolev and Leisner, 2020). SIP through rime-splintering is unlikely as it would have reduced the available $N_d$. This hypothesis is in line with the findings of Pasquier et al. (2022a), where the effect of SIP was inferred in about 40% of the in-cloud measurements.”

Comment:
L437: “…accumulation mode particles transported over long-range pathways...” This is the first point that I recall hearing of the source of the accumulation mode particles in the paper. The accumulation mode particle source information should be discussed earlier or this part of text should be removed from the conclusions.
Reply:
In addition to the abstract, we also had the following discussion in Section 4.3:
“Although anthropogenic pollution transported from lower latitudes during the Arctic haze period (late winter and spring; Rahn, 1981; Hirdman et al., 2010) controls the CCN and droplet population, only a fraction of it was activated to cloud droplets, as larger $D_{act}$ values, centered around 50-100 nm, were in the range of accumulation mode particles typically linked to this type of atmospheric transport.”
No change was made.

Comment:
L449: “This also means that warm cloud activation theory, such as described by well-established activation parameterizations (e.g., Morales and Nenes, 2014) are appropriate for application in mixed-phase cloud simulations.” Again, given uncertainty in how representative the balloon data are, this might be overstating the findings.
Reply:
We kindly disagree, as we are not saying that droplet number is always equal to that from warm cloud theory, but rather that the process of droplet formation is well described by it. There could indeed be situations (outside of the observation period) where activated droplet number is decreased from collision-coalescence, riming, entrainment, etc., but that is beyond the scope of this statement. Nevertheless, in response to the reviewer’s point, we made the following changes:
This also argues that warm cloud activation theory, such as described by well-established activation parameterizations (e.g., Morales and Nenes, 2014) may be appropriate for application in mixed-phase cloud simulations.

Comment:
L69: “allowed us to conclude”
Reply:
This was corrected.

Comment:
The abbreviations ToF-ASCM, TOF-ASCM, and ACSM were all used to describe the same instrument. Please use one abbreviation consistently throughout the manuscript.
Reply:
This was corrected.

Comment:
Caption, Fig. 3: I found the first sentence to be very confusing. Please consider re-wording and making it into several separate sentences for clarity. For example, this is how I rearranged the information so I could understand it: “Predicted potential droplet number concentration ($N_d$) compared to measured integrated particle number concentration ($Naer$). Data are shown in 12 panels corresponding to the four seasons and an assumed updraft velocity ($\sigma_w$) of either 0.1, 0.2 or 0.3 m s$^{-1}$. $N_d$ is an output of the cloud droplet formation parameterization. $Naer$ is measured by the two DMPS instruments.”
Reply:
This was corrected following the proposition of the reviewer.

Referee #2 has raised important questions concerning key points of the manuscript. We would like to thank them for the work they produced. The modifications they proposed were taken into account as detailed below.

Comment:
1 - A weakness of the current study is that CCN were not directly measured. The chemical composition was used to predict activation from the aerosol size distribution, but as shown in Figure S3, two different estimates didn’t agree that well. This uncertainty was treated via a sensitivity study which found that overall, the hygroscopicity was a second-order effect (line 353). This isn’t surprising, given the dependencies on size and composition that are seen in the Kohler equation, which shows that the aerosol size distribution is the more important factor. The maximum size measured by the SMPS was 945 nm (for some periods). Can the authors comment on whether a larger mode was ever present? At some low updraft speeds, the presence of larger particles might play a role in scavenging vapor and modifying Smax. An alternative sensitivity study might be to assume k=0.3 (where the data in Figure S3 appear to cluster and which is frequently selected as representative) and to use uncertainty in aerosol size and/or presence of a larger mode as the variable to be tested.

Reply:
This is an interesting point raised by Reviewer #2.
DMPS measurements at Zeppelin throughout the year 2020 are shown below (not shown in the manuscript):

![DMPS measurements plot](image)

This plot indicates that the mode diameter oscillated between approximately 30 and 300 nm throughout the year, with very few particles larger than 500 nm. We thus consider it very unlikely that a second mode larger than 900 nm could exist. Previous literature in which particle size distribution was measured at Zeppelin, either with a DMPS, with an aerosol particle sizer (APS; measuring the size distribution of particles larger than 500 nm), or from a PALAS-FIDAS (fine dust measurement device) optical counter did not show any additional mode, and supermicron particles represented only very minor fraction of aerosol population (Tunved et al., 2013; Pasquier et al., 2022b).

We included the following text in Section 4.3:
The presence of very large particles, larger than the maximum diameter that can be detected by the DMPSs, could potentially scavenge water vapour and cause drops in $S_{\text{max}}$ as low as those we observed. However, the mode diameter measured during the campaign was consistently between about 30 and 300 nm, and particles larger than 500 nm were very rare (not shown), in agreement with previous literature at the same site (Tunved et al., 2013; Pasquier et al., 2022b). We thus consider it very unlikely that low values of $S_{\text{max}}$ are caused by this phenomenon.

Comment:
The updraft velocity comparisons shown in Figures S1 and S2 raise some questions. The wind lidar seems to have a very small range of observations compared to the other measurements and the uncertainties appear to be large. This is discussed some in Section 3.2, but it wasn’t clear how much the results in figure 3 or the degree of “closure” are affected. Does it imply that, for Figure 2, most of the variability is driven by the variability in aerosol number concentrations? Comparing Figure 1a, this seems plausible.

Reply:
As shown in Figure S2, the comparison between uSonic- and wind LiDAR-derived updraft velocity provides relatively accurate but not very precise results. This could be due to fine-scale variability in vertical motion but also to larger-scale differences (as mentioned in the manuscript, the two instruments were separated by about 2 kilometres in horizontal distance). However, both instruments appear to have a similar range of observation, as shown by the circular shape of the cloud of points in Figure S2 but also in Figure S1 when northern wind-influenced data points are removed (they are influenced by local topography).

We believe that the variability in Figure 2 is indeed largely caused by the variability in aerosol number concentration, as indicated by the comparison between Figures 2b (now moved to Figure S4c) and 1a, but mostly by the near-linearity seen in every panel of Figure 3.

Figure 2 (the new Figure S4) gives a good indication of the second order influence of the updraft velocity on the outputs of the cloud droplet formation parameterization: the results based on the wind LiDAR and on the uSonic strongly agree.

The following paragraph was added to Section 4.3:
“The comparison between uSonic- and wind LiDAR-derived updraft velocity shown in Figure S1 and S2 provides relatively accurate but not very precise results, which could be due to fine-scale variability in vertical motion but also to larger-scale differences related to the horizontal distance between both instruments. However, Figs. 2 and S4 provide a good indication of the second order influence of the updraft velocity on the outputs of the cloud droplet formation parameterization, since the $N_d$, $S_{\text{max}}$ and $D_{\text{act}}$ results based on the wind LiDAR and on the uSonic strongly agree.”

Comment:
Section 4.5: it would be appropriate to note that closure was assessed to be attained when predictions were within a factor of 2 of observations. Also, there are only two cases for comparison which should be made more clear in the Abstract.

Reply:
The criterion for closure validity was added to the following sentence:
“The closure, which is assessed to be attained when predictions were within a factor 2 of observations, appears successful for $N_d > 8-10$ cm$^{-3}$, thereby validating the use of the cloud droplet formation parameterization in the Arctic environment.”

The abstract was modified following a similar comment from Reviewer #1:
“We evaluate the parameterization and the droplet numbers calculated through a droplet closure with in-cloud situ measurements taken during 9 flights over 4 days. A remarkable finding is that, for the clouds sampled in situ, closure is successful in mixed-phase cloud conditions regardless of the cloud glaciation fraction.”

Comment:
Section 4.5: For the two cloud cases, were these single layer clouds? There should be some more discussion of the cloud characteristics that helps show the comparison is appropriate.

Reply:
Section 4.5 directs to the study of Pasquier et al. (2022) for detailed descriptions of the atmospheric conditions during the flights. Looking at their Figure 3, the clouds cases during the first 3 days (10-12 November 2019) were single-layered clouds, but the day in April is a typical seeder-feeder configuration (there is a synoptic cloud above). We modified the text as follows:
“This was achieved several times in past studies through successful droplet closures (e.g. Fountoukis et al., 2007; Kacarab et al., 2020), and can also be performed here using measurements from the HOLIMO taken on 9 Holoballoon flights during the NASCEN'T campaign (Fig. 5; see Pasquier et al., 2022a) for detailed descriptions of the atmospheric conditions during the flights. We note that, based on the radar measurements they performed, the clouds studied in November 10, 11 and 12, 2019 were monolayer clouds, but the April 1$^{st}$ case is a typical seeder-feeder configuration, with a synoptic cloud above the sampled cloud).”

Comment:
I found this statement a little hard to follow; perhaps the authors can rework this a bit.

Reply:
This paragraph now reads as follows:
• Several recently published studies focusing on the factors limiting cloud droplet formation were able to distinguish periods of aerosol-limited or updraft velocity-limited cloud droplet formation regimes in boundary layer clouds with high updraft velocities such as cumulus and stratocumulus clouds, but also in alpine mixed-phase clouds that can form in more stable air dynamics. The current study demonstrates that updraft-velocity cloud formation can also occur in a relatively unpolluted environment with weak convection of maritime air masses such as the Arctic, during winter and early spring.

Comment:
did the DMPS measurements use impactors on the inlets?

Reply:
The DMPS systems did not use any impactor.
We modified the text as follows:
“Condensation particle counters (CPC, TSI models 3010 for DMPS_1 and 3772 for DMPS_2) then measure the concentration of particles contained in the monodisperse flow. No particle impactor was used.”

References


