

We thank the editor and reviewers for the handling and commenting of our manuscript. Please find below our point-by-point responses to each of the reviewers' comments, including the modifications we have made to the manuscript. Reviewer comments (RC) are in black text and the answers to reviewers (AR) are given in *blue text*, and excerpts of the revised manuscript are given in *blue italic text*.

#### **Reviewer 1 :**

The study by Pohorsky et al. provides valuable new insights into aerosol-cloud-radiation interactions in the Arctic, based on tethered balloon measurements above the sea ice. Profile observations of meteorological parameters, as well as aerosol and cloud properties, were effectively combined with cloud remote sensing and modeling to investigate the impact of free-tropospheric aerosols on low-level cloud properties. The authors showed that cloud droplet numbers could not be explained solely by boundary-layer aerosol concentrations but required the inclusion of the entrainment of cloud condensation nuclei (CCN) at cloud top. Radiative transfer modelling resulted in an enhancement of the cloud's longwave warming by 1.3 W m<sup>-2</sup> by the entrained particles. This effect is of the same order as previously reported effects of anthropogenic pollution on Arctic clouds and might be even more pronounced under conditions with fewer aerosols. Although the study is based on a single case, it provides an important piece in assessing the CCN budget of Arctic low-level clouds and their radiative effects. Moreover, tethered-balloon observations over Arctic sea ice are sparse due to the inherent difficulties of conducting them. The paper is of high scientific quality, is well-structured, and reads well. I recommend accepting the paper with minor revisions.

*We would like to thank the reviewer for the positive and constructive comments on the manuscript. Answers to individual comments are provided below.*

RC1: One major topic that needs to be treated in more detail is the cloud droplet measurements and their use in the models. The study would be significantly strengthened if the LOAC cloud droplet size distributions were used to derive the LWC and effective radius  $r_{\text{eff}}$ , rather than remote-sensing products. Or at least should be compared with remote sensing, as the droplet number concentration profiles in Fig. 7 indicate a smaller  $r_{\text{eff}}$  at the bottom of the cloud than in the upper part.

*AC1: We agree that cloud microphysical properties derived directly from the LOAC measurements provide a valuable complement to the remote-sensing products. Our original choice to use the remote-sensing retrievals was motivated primarily by their continuous temporal coverage throughout the day, whereas the LOAC measurements were only available during balloon profiles and were occasionally affected by detector saturation at high droplet concentrations, resulting in data gaps.*

*Prior to the radiative transfer calculations, we compared the liquid water content (LWC) derived from the LOAC measurements with the remote-sensing retrievals and found very good agreement between the two approaches. A figure has been added to the SI to illustrate this correspondence (Fig. S5). Since the radiative transfer model requires LWC and cloud effective radius as inputs, and since the remote-sensing products provide continuous observations of these quantities throughout the cloud lifecycle, we consider them the most appropriate dataset for the purposes of this study.*

*The objective of this work is to assess the sensitivity of cloud microphysical and radiative properties to aerosol-related assumptions while keeping the cloud state constrained by observations representative of those typically available in atmospheric models and observation-based studies. In such applications, cloud properties are generally derived from continuous ground-based remote-sensing observations rather than*

from in situ cloud microphysical measurements. The use of remote-sensing-derived LWC and effective radius is therefore consistent with the intended framework of the study.

Because the LOAC-derived and remote-sensing-derived cloud properties were found to be in good agreement, and because our focus is not on evaluating differences between cloud microphysical datasets but rather on isolating the effects of aerosol assumptions, we did not consider additional radiative-transfer simulations based on LOAC-derived cloud properties to be necessary.

L301-305: *The remote-sensing retrievals were used because they provide continuous cloud microphysical information throughout the day, whereas the LOAC measurements were only available during balloon profiles and occasionally contained data gaps caused by detector saturation at high droplet concentrations. To evaluate the consistency between both approaches, LWC derived from the LOAC droplet size distributions was compared with the remote-sensing retrievals and showed good agreement (Fig. S5). Details on LWC and  $r_{eff}$  calculation from the LOAC measurements are provided in the SI.*

SI L58-64: *From the LOAC measurements, the liquid water content was calculated as:*

$$LWC = \sum_{i=1}^n N_i \frac{4}{3} \pi \left(\frac{D_i}{2}\right)^3 \rho_w 10^{-6}, \quad (1)$$

where  $i$  represents the size bin and  $n$  the number of size bins.  $N_i$  is the droplet number concentration for bin  $i$ ,  $D_i$  is the midpoint diameter of droplets in bin  $i$  (in  $\mu\text{m}$ ), and  $\rho_w$  is the density of water ( $1 \text{ g cm}^{-3}$ ). The LWC is expressed in  $\text{g m}^{-3}$ .

The corresponding effective radius was calculated as

$$r_{eff} = \frac{\int_0^{\infty} r_i^3 n(r_i) dr}{\int_0^{\infty} r_i^2 n(r_i) dr}, \quad (2)$$

where  $r$  is the droplet radius associated with size bin  $i$ .

RC2: It would also be beneficial to add the cloud droplet number distribution as an average over the cloud layer, or, better yet, as a vertical contour plot, rather than droplet number concentration per-bin profiles.

AC2: We thank the reviewer for this suggestion. We considered alternative representations of the droplet size distribution, including cloud-layer-averaged distributions and vertical contour plots. However, we have retained the original presentation of the droplet number concentration profiles for the individual size bins.

The purpose of this figure is to illustrate the vertical evolution of the droplet population through the cloud layer, in particular the increase in larger droplets toward cloud top and the corresponding variation in total droplet number concentration. The individual bin profiles provide a direct and transparent view of these features while preserving the vertical structure of the observations.

A cloud-layer average would remove the vertical information that is central to the analysis, and we do not believe that a contour representation would provide additional insight relevant to the objectives of this study. For this reason, we have chosen to retain the original figure.

RC3: The droplet number concentration of the LOAC should be validated in greater detail in the main part of the paper, since this is the core part of the study that connects different observations to the models, e.g., by performing a linear regression against the fog monitor.

AC3: A comparison between the LOAC and Fog monitor was performed at the end of the campaign where the two instruments measured side by side for a period of almost two days with two main fog events. A qualitative comparison was already shown in Fig. S3 and discussed in Sect. 2.2.

We agree that a more complete validation of the LOAC data is important. We have therefore modified Fig. S3 to provide a more complete comparison and discuss the comparison in the SI to keep the method section more concise.

The main text now reads as follows:

L195-198: *A comparison of the data from the LOAC and a fog monitor (FM120, Droplet Measurement Technologies, USA) shows that the LOAC droplet number concentration agrees within 20 to 30% with the reference instrument. Details of the analysis are shown in the SI (Fig. S3).*

And the following description was added to the SI along with the new Fig. S3.

SI L42-48: *A quantitative comparison between the LOAC and a reference instrument (FM120, Droplet Measurement Technologies, USA) was performed during a side-by-side deployment at the end of the campaign covering two fog events (Fig. S3). The droplet concentrations measured by the two instruments were well correlated (adjusted  $R^2 = 0.83$ ), indicating that the LOAC successfully captured temporal variations in cloud droplet number concentration. A linear regression yielded a slope of 0.74, suggesting that the LOAC systematically reported lower droplet concentrations than the FM120. Given the scatter in the comparison, the LOAC-derived droplet concentrations should be considered accurate within approximately 20–30 %.*

RC4: In section 2.2, it is stated that  $N_d$  is derived from droplets larger than  $3 \mu\text{m}$ , which is inconsistent with Fig. 7 where smaller sizes are included. On what size range are the reported  $N_d$  of  $130 \text{ cm}^{-3}$  in the case study based on?

AC4: Thank you for spotting this inconsistency.  $N_d$  was indeed calculated for “particles” larger than  $3 \mu\text{m}$  but we kept all the bin sizes for the figure. We corrected the figure to be consistent with our analysis and removed size bins below  $3 \mu\text{m}$ .

RC5: One aspect that puzzles me is why the case study is based only on one of the six available profiles on 7 June? Including the other profiles would significantly strengthen the study by enhancing the statistics or providing more insights into the variations between the profiles.

AC5: The analysis was not restricted to a single profile. The profile shown in Fig. 7 was included as a representative example to illustrate the vertical structure of the atmosphere and the cloud. Displaying all six profiles would have resulted in substantial redundancy, as they exhibited similar overall characteristics,

while differences in cloud-base and cloud-top altitude between profiles would have made the presentation of an average profile difficult to interpret.

The aerosol size distribution and cloud microphysical analyses were performed using data from all six profiles collected on 7 June. Thus, the results presented in the overall analysis (including the PNSD analysis from Fig. 8) are based on a larger dataset rather than on the single profile shown in Fig. 7. We have clarified this point in the revised manuscript.

*L454-457: Figure 7 shows a representative snapshot of the atmospheric column during the third ascent of the day (from 19:09 to 20:06 UTC). This profile is presented to illustrate the vertical structure of the atmosphere, cloud, and aerosol layers. Unless otherwise stated, the subsequent aerosol size distribution and cloud microphysical analyses are based on data from all six profiles obtained on 7 June.*

We would also like to point out that no other flights through clouds during the campaign contained such a complete dataset or such stable conditions, which is why our analysis focused on this case study.

**RC6:** Judging from Fig. 7, it seems that the aerosol particle number size distribution of the entrainment zone is only based on a single mSEMS scan at ~400 m height. This provides rather weak statistics, given the counting statistics of small charged particles at low aerosol concentrations and an mSEMS scan time of 160 s during an ascending balloon flight in a dynamic environment.

**AC6:** The aerosol particle number size distributions shown in Fig. 8 were not derived from the single profile displayed in Fig. 7. Fig. 7 is included only as a representative illustration of the vertical structure of the atmospheric column. The size distributions presented in Fig. 8 were computed using data from all six balloon profiles. For the entrainment zone, this corresponds to a total of eight mSEMS scans. Although individual scans are subject to statistical noise, the overall shape of the size distribution was consistent across the scans, providing confidence that the observed features are representative.

Detailed comments:

**RC7:** Figure 1: Nice figure, but also slightly complex with the particle sources in the free troposphere (long-range, NPF) not mentioned in the introduction so far. Better focus on the core points of the study in the figure, or give background on the particle sources in the intro, including references.

**AC7:** We agree that the purpose of Fig. 1 is to illustrate the conceptual framework of the study rather than to provide a comprehensive overview of all possible aerosol sources in the Arctic free troposphere. Since the origin of the aerosol layers aloft is not a primary focus of this work, we simplified the figure by removing the explicit references to long-range transport and new particle formation (NPF). The revised figure now focuses on the key processes investigated in this study, namely cloud-top entrainment and the contrast between cloud seeding by boundary-layer aerosols only and cloud seeding including aerosols from above the cloud.

**RC8:** Sec 2.1: What was the average climb rate of the balloon and the resulting vertical resolution of the mSEMS?

**AC8:** The average vertical velocity of the Helikite during continuous ascents and descents was approximately  $15 \text{ m min}^{-1}$ , although it varied depending on wind conditions. Given the mSEMS scan duration of 2 min 40 s, a single scan corresponds to an average vertical distance of approximately 40 m.

To improve counting statistics, the Helikite was frequently held at selected altitudes during descents, resulting in longer sampling periods than would be obtained during a continuous profile. For the case study presented here, hovering periods were systematically performed at key levels, including above the cloud, within the cloud, and below cloud base (e.g., Fig. 6).

We have added the following information to Sect. 2.1:

L140-141: The maximum altitude reached was 645 m above sea level *and the average vertical velocity of the balloon during operations was  $\approx 15 \text{ m min}^{-1}$ .*

L142-145: A miniaturized scanning electrical mobility spectrometer (mSEMS model 9404, Brechtel Manufacturing Inc., USA) provided particle number size distribution (PNSD) measurements for particles with electrical mobility diameters between 8 and 280 nm with 40 log-spaced size bins and a bin time of 4 seconds, yielding a time resolution of 2 min 40 sec per scan, *which corresponds to  $\approx 40$  meters on average during ascents and descents of the Helikite.*

RC9: L 157–161: This could be moved to the supplementary material, as the STAP and filter samples were not used in the paper.

AC9: We agree. The paragraph has been moved to the SI after Table S1.

RC10: Supp. Fig S1: Typo in caption, d) N8-280

AC10: Thank you, the typo has been corrected.

RC11: L. 186 -191: The POPS correction scheme seems inaccurate. It seems more reasonable to correct the POPS diameter rather than the particle number to match the DMPS. The particle numbers of the POPS are probably correct, but they are counted at a different size due to the deviation of the optical diameter from the mobility diameter. This would also affect the lower detection limit of the POPS.

AC11: We agree that, in principle, a correction based on converting optical diameters measured by the POPS to mobility diameters measured by the DMPS would be physically more rigorous than correcting particle number concentrations. However, such a conversion requires knowledge of aerosol properties such as the complex refractive index and particle shape, which were not available for the aerosol population investigated here. Furthermore, the relationship between optical and mobility diameter is generally size-dependent and cannot be represented by a simple uniform diameter shift across the entire size range.

Given these limitations, we adopted an empirical correction based on the observed size-resolved differences between the POPS and DMPS measurements. While this approach does not explicitly account for the underlying diameter conversion, it provides a practical means of reconciling the two datasets without introducing additional assumptions regarding aerosol optical and morphological properties. Because the analysis focuses on particles within the upper part of the accumulation mode, where the POPS and DMPS size distributions show overall good agreement after correction, we expect the remaining uncertainty associated with the chosen correction approach to be small relative to the other sources of measurement uncertainty.

RC12: Fig. S3: What are the lower Y-Axis labels on the heat maps, droplet size?

AC12: Yes, that is correct. The axis labels have been modified to clarify the information.

RC13: L 233: Unintroduced abbreviation SLP

AC13: The acronym is now correctly introduced.

RC14: Fig S4: Typo in caption: 186 and 3370 nm

AC14: Thank you, the typo at line 42 has been corrected.

RC15: Sec. 2.6: The droplet activation scheme seems to be derived for liquid stratus clouds. Is it applicable to mixed-phase clouds, as in your case study?

AC15: The reviewer raises an important point. The cloud droplet activation parameterization was originally developed to predict activation in warm clouds and does not explicitly represent mixed-phase cloud processes. However, in this study the parameterization is used only to estimate the number of droplets activated from the observed aerosol population and updraft velocity. This activation process remains governed by CCN activation physics and is therefore applicable irrespective of whether the cloud subsequently evolves as a liquid or mixed-phase cloud. Furthermore, the cloud investigated here was liquid-dominated, with cloud-base temperatures around  $-5\text{ }^{\circ}\text{C}$  and observed cloud droplet concentrations substantially exceeding ice crystal concentrations.

The same parametrization has previously been used by Motos et al. (2023) for Arctic low-level clouds in Ny-Alesund and demonstrated closure between the parametrization and in situ measurements even for mixed-phase clouds, regardless of the glaciation fraction. Hence, we are confident that the parametrization is applicable to our case study.

RC16: L 382-383: The number  $\sim 150\text{ cm}^{-3}$  is inconsistent with  $\sim 450\text{ cm}^{-3}$  on 10 June, as seen in Fig. 4. The statement “– too small to activate in this case” without the supporting PNSD seems difficult to comprehend.

AC16: The value of approximately  $150\text{ cm}^{-3}$  refers to the concentration of interstitial (i.e. non-activated) aerosol particles within the cloud and corresponds to the blue curve in Fig. 4 between cloud base and cloud top. The value of approximately  $450\text{ cm}^{-3}$  visible on 10 June represents the total aerosol concentration and therefore includes both activated and non-activated particles.

Our interpretation is that the relatively high concentration of interstitial particles on 10 June compared to the other cases is consistent with an enhanced contribution from nucleation- and Aitken-mode particles. Because these particles remained interstitial within the cloud, they were likely below the activation size under the supersaturation conditions encountered during this flight. We agree that this interpretation is not directly supported by the data shown in the manuscript and have therefore clarified the wording in the revised text as follows:

*L390-392: For the June 10 flight, the concentration of interstitial aerosols ( $\approx 150\text{ cm}^{-3}$ ) was greater than on other days consistent with increased concentrations of Aitken- and nucleation-mode particles, which likely remained below the activation size under the cloud conditions encountered during this flight.*

RC17: Fig 4: Please provide further info on how the mSEMS and POPS were merged in terms of bin limits and different time resolutions of each instrument to derive profiles of N8-3370.

AC17: For each mSEMS scan, the higher temporal resolution POPS data were averaged over the corresponding mSEMS sampling interval. The total aerosol number concentration ( $N_{8-3370}$ ) was then calculated by combining the mSEMS size distribution between 8 nm and the lower detection limit of the

POPS (186 nm) with the POPS size distribution above this diameter. The merged distribution therefore covers the size range from 8 nm (mSEMS lower detection limit) to 3370 nm (POPS upper detection limit).

This information has been added to the methods section.

L189-193: *To derive the total aerosol number concentration ( $N_{8-3370}$ ), the mSEMS and POPS measurements were merged. For each mSEMS measurement, the higher temporal resolution POPS data were averaged over the corresponding mSEMS sampling interval. The merged size distribution consisted of the mSEMS measurements between 8 nm and the lower detection limit of the POPS (186 nm), combined with the POPS size distribution above this diameter, resulting in a total size range from 8 to 3370 nm.*

RC18: Fig 5: Typo in caption: 1000 km radius should be 100 km, as in the text?

AC18: Thank you, the typo has been corrected.

RC19: L 436-438: sentence reads a bit difficult. How is the increasing concentration above the cloud linked to decreasing concentrations at the surface?

AC19: We agree that the original wording was ambiguous and could be interpreted as implying a causal relationship between the increasing concentrations above the cloud and the decreasing concentrations at the surface. This was not our intention. The decrease in surface  $N_{186-3370}$  is possibly related to scavenging by snowfall, whereas the increase in concentrations above the cloud is a separate observation whose origin remains uncertain. We have revised the text accordingly:

L445-447: *In addition to the apparent depletion within the cloud due to cloud droplet formation, we observe a decrease in  $N_{186-3370}$  at the surface throughout the day, possibly due to scavenging by snowfall. At the same time, concentrations above the cloud increase, although the cause of this increase remains unclear.*

RC20: L 466 – 469: This conclusion may be better placed later in the text, since the aerosol PNSD was not introduced yet.

AC20: We thank the reviewer for this comment. We agree that the original text introduced a conclusion before presenting the aerosol size distribution analysis that supports it. To improve the logical flow, we have softened the statement in the description of Fig. 7 to indicate only that the observed cloud droplet concentrations suggest a possible contribution from additional aerosol sources or vertical transport processes. The stronger conclusion that boundary layer aerosol alone is insufficient to explain the observed droplet concentrations, and that free-tropospheric aerosol entrainment provides an additional source of CCN, has been moved to the later section where the PNSD analysis and the estimate of the aerosol concentration above the Hoppel minimum are introduced.

Modification at L476-479: *In the present context, the observed cloud droplet concentrations are relatively high compared to the aerosol concentrations measured below cloud. This suggests that additional aerosol sources or vertical transport processes may contribute to the cloud CCN population, a hypothesis that is explored in the following part through analysis of the aerosol size distributions.*

Main conclusion moved to:

L527-530: *These results indicate that the aerosol population within the boundary layer alone is insufficient to explain the observed cloud droplet concentrations. Therefore, an additional source of CCN*

is required. The observations in the entrainment zone and lower free troposphere suggest that entrainment of aerosol-rich air from above the cloud provides a plausible source of these additional CCN.

RC21: Fig 8: Please add the DMPS size distribution for the PBL to the plot, or add a comparison of the mSEMS with the DMPS to the supplementary.

AC21: We have added the average DMPS size distribution to Fig. 8 (blue dashed line, same color as boundary layer/ground mSEMS measurements) and completed the caption. The DMPS and mSEMS measurements show an excellent agreement.

RC22: L547 – 551: Does the calculated number of CCN above the Hoppel minimum from the DMPS match the measured number from the CCN counter, when it is derived by an interpolation between the two nearest SS levels, similar to the procedure for deriving kappa?

AC22: Yes, the agreement between the CCNC and DMPS measurements was thoroughly evaluated prior to the analysis and was found to be good. By interpolating the CCNC concentrations between the two nearest supersaturation levels (0.3 % and 0.5 %) to estimate the CCN concentration at 0.42 % supersaturation (derived from our  $\kappa$  calculations), we obtain values that generally agree with the integrated DMPS concentration above the Hoppel minimum within approximately 10 %, as shown in Fig. 1. This agreement provides confidence that the activation diameter inferred from the Hoppel minimum is consistent with the CCNC observations.

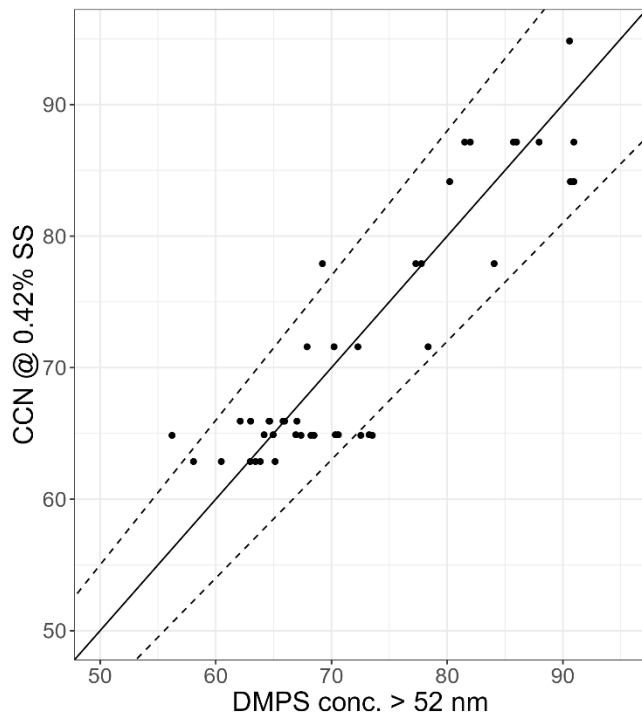


Figure 1 Scatterplot of interpolated CCN concentration at 0.42% and corresponding integrated aerosol concentration above the Hoppel minimum during the flight period on June 7, 2023.

RC23: L627 – 628: There seems to be a typo in “3 times more droplets”, as your range of Nd from 68 to 122 cm<sup>-3</sup> is more or less similar to the study.

AC23: The statement refers to the results of Garrett and Zhao (2006), not to the range of Nd observed in our case study. In their study, Nd increased from 53 cm<sup>-3</sup> under clean conditions to 153 cm<sup>-3</sup> under polluted conditions, corresponding to a factor of approximately 2.9, which is significantly more than the 1.8 increase of our study. To avoid ambiguity, we have reworded the sentence as follows:

*L642-644: While the LWP reported by Garrett and Zhao (2006) is similar to that of our case study, the increase in Nd between their clean and polluted cases was larger than in our observations, with Nd increasing from 53 to 153 cm<sup>-3</sup> (a factor of approximately 2.9).*

#### **Reviewer 2 :**

Review of “Contribution of free tropospheric aerosols to Arctic low-level cloud droplets formation and longwave radiative forcing” by R. Pohorsky et al.

This is a nice analysis of data from the recent ARTofMELT campaign to probe the importance of enhanced aerosol concentrations above cloud tops. The data analysis and the methodology is generally good, but I am struggling to make sense of the results and hopefully the authors can provide some clarification.

We would like to thank the reviewer for the constructive comments on the manuscript and hope the answers below will bring clarity.

#### Major Comments:

RC1: It is absolutely reasonable to think that the entrainment zone aerosol particles are contributing to the in-cloud droplet concentration. However, there is one aspect that I can't understand. The droplet concentration is well-mixed through the cloudy layer suggesting that particles activated at cloud top quickly become dispersed through the cloud. Wouldn't it follow that ALL particles, including the unactivated Aitken mode particles, would also become well-mixed throughout the cloudy and sub-cloud layers? And if that were true, shouldn't the sub-cloud aerosol population be sufficient to explain the droplet concentration? This statement should apply to the accumulation mode particles too since the cloud and sub-cloud layers are coupled. In other words, if you dried out the cloud droplets and counted accumulation mode particles, shouldn't it roughly match the sub-cloud population? But with the current explanation, I don't think this would be true. With the given explanation there would be a substantially higher number of accumulation particles in the cloud layer than in the sub-cloud layer even though the Aitken populations in these two layers match. I can't understand why one aerosol mode should match and the other mode shouldn't.

AC1: We thank the reviewer for this insightful comment. We agree that, if aerosol particles entrained from above cloud were rapidly and completely mixed throughout the cloud and sub-cloud layers and behaving as passive tracers, one might expect both the Aitken and accumulation modes to become homogenized and the sub-cloud aerosol population to be sufficient to explain the cloud droplet concentration.

However, several factors complicate this assumption. First, the droplet concentration is not perfectly homogeneous throughout the cloud layer, with somewhat larger concentrations observed toward cloud top. This points to a non-homogeneous vertical distribution of accumulation mode particles. Aerosol particles are also subject to activation and scavenging processes and cannot be treated as conserved tracers. For example, Aitken mode particles can be scavenged in droplets. Consequently, the vertical distributions of the different aerosol modes are not expected to evolve identically.

We observe, however, that at cloud top the size distribution is consistent with a mixture of the two aerosol populations (i.e. in-cloud and above cloud aerosol size-distribution), which is in line with the interpretation of entrainment of free-tropospheric aerosol into the cloud.

Because the available observations do not permit us to distinguish between these processes or quantify their relative importance, we cannot provide a definitive explanation for the differing behavior of the Aitken and accumulation modes. This is indeed an important question that requires further investigation.

We have therefore softened our interpretation and now state that the enhanced aerosol concentrations above cloud are consistent with a contribution from entrained aerosol to the cloud droplet population, rather than providing direct evidence that these particles are solely responsible for the observed droplet concentrations.

Text modification in Sect. 4.2

L479-480: *This suggests that additional aerosol sources or vertical transport processes may contribute to the in-cloud CCN population, a hypothesis that is explored in the following part through analysis of the aerosol size distributions.*

L527-530: *These results indicate that the aerosol population within the boundary layer alone is insufficient to explain the observed cloud droplet number concentrations. Therefore, an additional source of CCN is required. The observations in the entrainment zone and lower free troposphere suggest that entrainment of aerosol-rich air from above the cloud provides a plausible source of these additional CCN.*

Text modification in Sect. 5 (Conclusion)

L681-684: *...indicating mixing between cloud air and air from above the cloud, consistent with the entrainment of free-tropospheric aerosols into the cloud. The analysis of the cloud microphysical properties indicated that the aerosol particles measured in the boundary layer alone may not fully explain the observed cloud droplet concentrations, suggesting that an additional source of CCN associated with aerosol above the cloud likely contributed to droplet formation.*

L687-692: *Parametrizations including aerosols measured above the cloud in addition to those in the boundary layer produced approximately 80 % more cloud droplets than input based solely on boundary layer aerosols. The resulting cloud droplet concentrations were in substantially better agreement with the in situ observations, supporting the interpretation that aerosol entrained from above cloud contributed to the observed droplet population. Although the cloud appeared coupled to the surface, surface-based measurements alone may not fully represent the aerosol populations available near cloud top, and therefore may not adequately capture the influence of aerosol entrainment from above.*

L700-701: *...indicating that under such conditions the contribution of aerosol sources above the cloud may become increasingly important.*

L707-709: *These limitations should be considered when improving the representation of Arctic LLCs in models. In particular, aerosol sources aloft and the associated entrainment processes may need to be represented more explicitly.*

Text modification in Sect. 5 (Conclusion)

L27: *...consistent with entrainment of free-tropospheric aerosol.*

L29-30: *...improved agreement with observed droplet concentrations. Our observations through the cloud allowed us to highlight the potential importance of free-tropospheric CCN sources, ...*

RC2: I am confused by the droplet concentrations, and perhaps this confusion is related to my first comment. In Lines 192-193, the authors state that cloud droplets are those particles detected by the LOAC that are larger than 3 microns and classified as droplets. However, if Figure 7, size bins are shown for drop sizes of 0.9-1.1 microns and 1.1-3 microns. Are the concentrations of these smallest particles included in the total droplet concentration? I think the answer is “yes” but it is hard to tell given the log-scale used in Figure 7. Is this then why the droplet concentration is higher than the sub-cloud accumulation mode concentration? Perhaps the authors are including counts of hydrated but unactivated Aitken particles with sizes of 0.9-3 microns. Please check.

AC2: The reviewer correctly identified an inconsistency between the description of  $N_d$  in the text and the size bins displayed in Fig. 7.  $N_d$  was calculated exclusively from particles classified by the LOAC as water droplets and larger than 3  $\mu\text{m}$  in diameter. However, Fig. 7 originally displayed all LOAC size bins, including those below 3  $\mu\text{m}$ . To make the figure consistent with the analysis, we have removed the bins below 3  $\mu\text{m}$  from the revised figure.

We confirm that particles in the 0.9–1.1  $\mu\text{m}$  and 1.1–3  $\mu\text{m}$  size ranges were not included in the calculation of  $N_d$ . Regarding the possibility of hydrated but non-activated particles, only particles classified as water droplets by the LOAC algorithm were considered in the droplet concentration. Under cloud supersaturation conditions, particles growing to diameters above 3  $\mu\text{m}$  would be expected to have activated and formed cloud droplets.

RC3: It's not clear to me why the authors did not repeat the analysis for the other three cases identified. It seems that doing so could strengthen the conclusions if they are similar.

AC3: The objective of this study was to present a detailed case study of the impact of enhanced free-tropospheric aerosol concentrations on the microphysical and radiative properties of a low-level Arctic cloud. We agree that extending the full analysis to the other identified cases could provide additional insight. However, the selected case offered the most complete and internally consistent dataset for such an analysis. In particular, the cloud persisted throughout the day with relatively stable thermodynamic properties, continuous cloud cover, and comprehensive aerosol and cloud microphysical measurements.

In contrast, the other cases were characterized by more heterogeneous cloud conditions, including partial cloud cover and greater temporal variability. In addition, some flights lacked reliable cloud droplet size distribution measurements due to the absence or suboptimal performance of the LOAC instrument. As a result, the key variables required for a quantitative assessment of aerosol-cloud-radiation interactions were not consistently available across all cases.

The purpose of presenting all five cloud-top profiles was therefore to demonstrate that enhanced aerosol concentrations above the cloud were not unique to the selected case, but rather a recurrent feature observed during the campaign. The detailed analysis was restricted to the case for which the most complete and robust dataset was available.

Minor Comments:

RC4: Lines 437-438: I don't understand why increasing concentrations above cloud top should help to explain the decreasing concentrations at the surface.

AC19: The same comment was made by reviewer 1.

We agree that the original wording was ambiguous and could be interpreted as implying a causal relationship between the increasing concentrations above the cloud and the decreasing concentrations at the surface. This was not our intention. The decrease in surface  $N_{186-3370}$  is possibly related to scavenging by snowfall, whereas the increase in concentrations above the cloud is a separate observation whose origin remains uncertain. We have revised the text accordingly:

L437-439: *In addition to the apparent depletion within the cloud due to cloud droplet formation, we observe a decrease in  $N_{186-3370}$  at the surface throughout the day, possibly due to scavenging by snowfall. At the same time, concentrations above the cloud increase, although the cause of this increase remains unclear.*

RC5: Consider showing the Aitken mode concentration profile in Figure 7 especially since particles larger than 52nm contribute to the droplet population and you discuss the Aitken mode in more detail later.

AC5: We thank the reviewer for this suggestion and agree that seeing the vertical Aitken mode profile could help visualize vertical mixing of aerosols.

However, we have chosen not to include a dedicated Aitken mode profile in Fig. 7 for several reasons. First, the Aitken mode upper limit (located at or around the Hoppel minimum) in this situation is not introduced until the later analysis of the size distributions, where the Hoppel minimum is explicitly identified and discussed. Introducing a mode-based separation in Fig. 7 would therefore require applying an a priori size cutoff that is not yet established.

Second, Fig. 7 is intended to present the full measured size-resolved aerosol structure from both instruments, preserving the highest available vertical resolution of the POPS observations together with the mSEMS. Introducing an additional derived Aitken-mode quantity would either complicate the figure or require to add a merged data product from the POPS and mSEMS above 52 nm to obtain a complete picture of the total aerosol concentration profile. Such a product would have a lower spatial resolution than the POPS trace that is currently plotted.

We therefore prefer to retain the current representation in Fig. 7 and introduce the discussion of Aitken- and accumulation-mode partitioning after this part, where the modal structure and its vertical distribution are explicitly discussed.

RC6: Consider also citing Sterzinger and Igel 2024: <https://acp.copernicus.org/articles/24/3529/2024/>

AC6: Done. Citation added in the introduction at line 61.