

We sincerely thank reviewer #1 for carefully reading our manuscript, and for their review and constructive comments. We have reviewed the comments and have revised the manuscript accordingly. Our response is given in a point-by-point manner below. Reviewer comment (RC) and authors answer (AA).

*In the study “Influence of air mass origin on microphysical properties of low-level clouds in a subarctic environment” by Doulgeris et al. microphysical cloud properties measured during eight Pallas Cloud Experiments in the Finnish subarctic region are analyzed with respect to their air mass origin based on the Lagrangian particle dispersion model FLEXPART.*

*The scientific approach is valid and the manuscript is structured in a clear and concise way.*

*However, two main deficits regarding the scientific relevance and thus the scientific quality of the study as described in the general comments would require major revisions.*

### **General comments**

RC1: *“1. Scientific relevance. The study is based on a large time series of measurements campaigns that have been conducted in a subarctic mostly pristine region adequate for the analysis of aerosol cloud-interactions (ACI). A clear statement is missing on how the presented results may advance the current state of the art. The dependence of cloud microphysics on the air mass origin (Twomey effect in continental air masses vs. marine air masses) and that cloud droplets are prone to grow in warmer air is known already from other studies (cited in the manuscript 1.346). Also, the introduction is not clearly leading to a research hypothesis or research question. Modifications in the Introduction and discussion of results as well as in the abstract and conclusions are required to specify the scientific relevance of the study in the context of existing literature. The identification and further interpretation of results that add new findings to the existing body of knowledge would be helpful for the ACI community and further studies. “*

AA1: We have modified the text to be clearer with the main goals and significance of this work. Cloud processes are considered an important component of climate change in the Arctic region (Wendisch et al., 2019). However, even though there is an increased demand for long term continuous ground based in-situ cloud measurements, unfortunately there is limited instrumentation to cover such demand. The atmospheric in-situ measurements community (in our case the European Research Infrastructure for the observation of Aerosol, Clouds and Trace Gases, ACTRIS) has identified the cloud droplet probes with surface installation as a potential method for continuous cloud in situ measurements (ACTRIS-PPP Deliverable D5.1: Documentation on technical concepts and requirements for ACTRIS Observational Platforms). However, measurements in conditions like those at our sub-Arctic location are very challenging. As a result, the dependence of cloud microphysics on the air mass origin in a subarctic mostly pristine region were rarely seen until now using such an in situ long term dataset. We agree with the reviewer that there were already excellent studies that investigates cloud microphysics and their connection to air mass origin (e.g., Fuchs et al. 2017; Cho et al., 2021). However, one of the main differences with those studies is the methodology used. E.g., Fuchs et al. (2017) explain the importance of air mass origin and its characteristics to cloud properties using satellite data. Cho et al. (2021) investigate the relationship of cloud properties and radiative effects with air mass origin during the

winter (from November to February, 2016–2020) at Ny-Ålesund, Svalbard, by means of a remote sensing approach using a combination of cloud radar, ceilometer, and microwave radiometer measurements. To our knowledge, this is the first study that connects extensive in situ cloud measurements to air mass origin. In this work, we point out that there is need to consider not only local meteorological parameters but also the air mass origin in investigations of cloud processes. The PaCE measuring period (during autumn) is crucial as it is a unique opportunity for both experiencing Arctic pristine air masses (Pernov et al., 2022) and being able to measure them in situ with ground-based cloud instrumentation. Moreover, the procedure of distinguishing cases that corresponds to a single air mass origin and not a mixed one is complicated and requires a huge amount of continuous data. In this work, in situ cloud data with ground-based cloud spectrometers from eight different autumn campaigns were obtained (2004 hours of cloud observations resulted in 706 hours of cloud observations that related to all clean air mass origins). From this dataset, the relationship between  $N_c$  and droplet size (i.e., the Twomey effect) was characterized for the different source regions. We proved that cloud microphysical properties and particularly the number concentration of cloud droplets have a strong dependence on the air mass origin. Using those findings, the ACI community can focus on further studies to investigate how aerosol and meteorology of different airmasses along with local meteorological parameters change the cloud microphysics and to what scale.

Some of the major changes were applied to abstract, line 22

“...Local). We observed clear differences in the cloud microphysical properties for the air mass source regions. Arctic air masses were characterized by low liquid water content (LWC), low cloud droplet number concentration ( $N_c$ ), and comparatively large median volume and effective droplet diameter. Western region (marine North Atlantic) differed from Arctic by both higher  $N_c$  and LWC. Eastern region (continental Eurasia) had only a little higher LWC than Arctic, but substantially higher  $N_c$  and smaller droplet diameter. Southern region (continental Europe) had high  $N_c$  and LWC, and very similar droplet diameter to the Eastern region. Finally, the relationship between  $N_c$  and droplet size (i.e., the Twomey effect) was characterized for the different source regions, indicating that all region clouds were sensitive to increases in  $N_c$ .”

To introduction, line 92 “...at Pallas. To our knowledge, this is the first study that connects extensive in situ cloud measurements to air mass origin. During autumn, clean, natural Arctic background conditions are significantly increasing (Pernov et al., 2022). Subsequently, this allows us to focus in this work on quantifying the impact of air mass origin (e.g., clean arctic vs. long-range transported air from continental Europe) on the microphysical properties of low-level clouds and their patterns based on measurements at the Pallas GAW station. To our knowledge, this is the first study that connects extensive in situ cloud measurements to air mass origin.”

To results, line 285 “(a marine environment that the natural Arctic background conditions are significantly increasing (Pernov et al., 2022))”

Line 321 “Averaged temperatures at Sammaltunturi for each air mass were  $-3.1^\circ\text{C}$  (SD  $2.5^\circ\text{C}$ ),  $-2.2^\circ\text{C}$  (SD  $5.9^\circ\text{C}$ ),  $1.3^\circ\text{C}$  (SD  $3.9^\circ\text{C}$ ) and  $-2.8^\circ\text{C}$  (SD  $2.01^\circ\text{C}$ ) for the arctic, eastern, southern and western region respectively. Furthermore, in all regions, there was no clear indication that there was any trend in  $N_c$  through different years of PaCEs.”

To summary and conclusions line 475, 478 and 488

“This result suggests that clouds occurrence depended on the different meteorological conditions that were associated with the different air parcels”

“The lowest values of cloud droplet concentration were related to clean arctic airmasses. According to theoretical considerations (Brenquier 1991; Pawlowska et al., 2006), the measurements of cloud droplet number concentration do not depend on the vertical position of the cloud spectrometer within the cloud layer.

“The above differences that were observed in cloud microphysical properties when the air masses were related to different regions show the need to investigate how the aerosol loading and meteorology of different airmasses along with local meteorological parameters change the cloud microphysics and in which scale.”

RC2:” 2. *Scientific approach. Cloud properties are analyzed according to their air mass origin in 5 predefined source regions. Cloud properties strongly depend on the air mass characteristics including humidity, wind speed, temperature etc. at different altitudes. Including air mass characteristics (e.g. from ERA5 reanalysis) in the analysis to understand differences in  $N_c$ , MVD, ED as started in Fig. 8 would make results more interpretable and scientifically relevant.*

*Also the approach of using predefined source regions is questioned as this classification may result in similar/mixed air mass characteristics as shown for the Eastern/Southern and Arctic/Western air masses in Fig. 8. More intuitive would be an automatic classification (grouping) based on the air-mass origins or pathways. “*

AA2:

Investigating cloud microphysical properties and revealing the main factors that are dependent and at what scale is a complex procedure and this is also highlighted in summary and conclusion section of the revised manuscript. In the revised manuscript, we include also meteorological information from each region as they were measured at the station. Our main scope in this work is to investigate low level clouds using in situ measurements and the influence of different air mass origin. We proved that the air mass origin significantly affects the number concentration of the cloud droplets (the aerosol loading from each region is expected to play a role in this case). When investigating the sizes of the cloud droplets, the sizes were influenced by the number concentration of the cloud droplets as suggested by the Twomey effect.

Although MVD is quite similar for the source region combinations as pointed out by the reviewer, the different source regions do stand out from each other when  $N_c$  and LWC are included in the comparison. The following figure and discussion were added in the supplementary material (SM) of the revised manuscript.

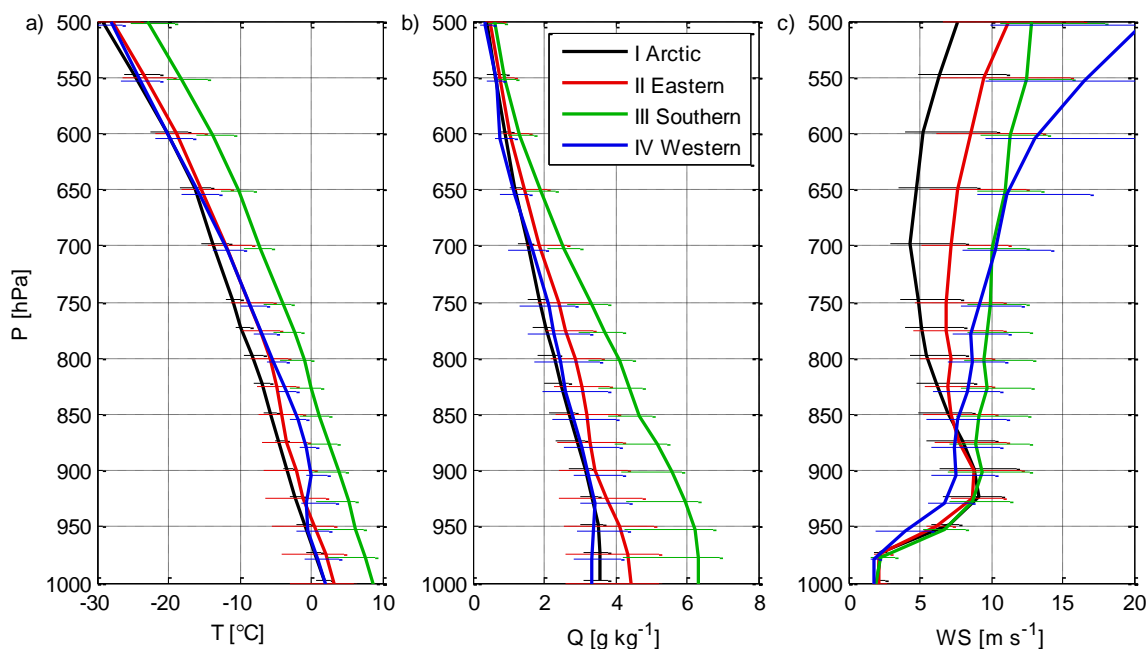


Figure S1. ERA5 temperature (T), specific humidity (Q) and wind speed (WS) profiles for the cases, when at least 80% of PES was within a source region. Line is the median and error bars indicate upper and lower quartiles. I, II, III and IV corresponds to the arctic, eastern, southern, and western region respectively. Station pressure is  $\sim 970$  hPa.

Temperature (T), specific humidity (Q) and wind speed (WS) profiles from ERA5 for the different source regions (Fig. 1) were compared. In ERA5 profiles, Southern source region stands out as the one with higher T and Q, which is also reflected in the observed cloud microphysical properties. For Western and Eastern region, the median profiles are quite similar to the Arctic profile, but the interquartile range is wider. For these source regions we observe higher variability in e.g., LWC compared to the Arctic source region, which suggests more variable meteorological conditions for these source regions. For WS the differences are relatively small.

We agree with the reviewer that finding the most important source areas is usually a difficult task in a variable environment and should be done with prudence. In the revised manuscript, we will elaborate on our decision to use the predefined regions to make clear to the reader the methodology that was used. The division of the areas is predefined as it was based on previous studies that were conducted at Sammaltunturi (e.g., Aalto et.al., 2003, Asmi et al., 2011). This work is a continuation of those studies and for this reason we decided to adopt the same 8 source areas. Initially, the regions were classified using trajectories cluster analysis, following the method as Eneroth et al. (2003) proposed. The predefined regions were used for different studies and scopes as atmospheric transport of carbon dioxide (Aalto et.al., 2003, Eneroth et al., 2005), aerosol studies (Tunved et al., 2006; Asmi et al., 2011). The choice of sectors represents roughly the characteristics of the region: the West and North are marine sectors, while the East and South are more continental sectors.

Line 224 "...Fig.3. The division was based on previous studies that were conducted at Sammaltunturi (e.g., Aalto et al., 2003; Eneroth et al., 2005; Tunved et al., 2005, Asmi et al., 2011). Initially, the regions were classified using trajectories cluster analysis, following the method that Eneroth et al. (2003) proposed. The choice of sectors represents roughly the characteristics of the region. The Arctic..."

### **Specific comments**

RC3: "*l. 106: Do you have information on the cloud type, is this mainly fog or low stratus? This may imply different processes.*"

AA3: The cloud type is low stratus or stratocumulus. This is diagnosed from the ceilometer observations: first, liquid cloud should be present at both the mountain top station and in the ceilometer profile for a minimum specified duration (30 minutes); and secondly, the liquid cloud base should be above the ground at the ceilometer location (the altitude of which is 210 m below the mountain top station). This ensures that we are not observing fog (at the ceilometer location) and that, since the cloud layer has to be present over a larger area to be included in a cloud event (present at both locations at the same time for a minimum duration of 30 minutes) and not varying much in height with time, we can then assume that it is stratus/stratocumulus at both locations.

RC4: "*l. 123: Latitude and longitude is missing in Fig. 1.*"

AA4: The above suggestion was accepted; A new map was created.

RC5: "*l. 201: Delete "model" as this can be mixed-up with numerical models.*"

AA5: The above suggestion was accepted.

RC6: "*l. 207: it is not specified if the PES belongs to an aerosol type or Nc or which emission inventory is used to calculate the PES. If solely air mass trajectories are calculated backwards what is the PES referred to? Please provide more details on the FLEXPART model settings and assumptions here.*"

AA6: Potential emission sensitivity (PES) is not connected to any emission inventory in this case, it is used only to characterize air mass history – therefore it is called “potential”. While PES can be clustered to retrieve air mass trajectories similar to e.g., Hysplit, we have chosen to utilize the PES field directly in the source region classification, as this accounts for turbulent mixing during the transport (c.f. Fig. 4 in the manuscript). Within FLEXPART, PES is calculated from a retroplume of inert tracers released at the measurement location and propagated backward in time. More information on PES can be found in Seibert and Frank (2004) and Pisso et al. (2019), which are referred to in the manuscript.

Lines 209-218 in the manuscript give already all the FLEXPART settings and describe the meteorological fields needed to re-run the simulations. Hence, we have made no changes to the manuscript.

“ERA5 reanalysis by European Centre for Medium-Range Weather Forecasts (ECMWF) was used as meteorological input fields for FLEXPART at 1 hour temporal resolution and 0.25° resolution in latitude and longitude. In vertical, ERA5 levels 50 to 137 were used, which corresponds approximately to the lowest 20 km above surface. The model domain was from 125° W to 75° E and 10° N to 85° N, which was large enough to contain 96 h simulations backward in time. FLEXPART runs were initiated at an hourly time resolution for the in-cloud measurement periods at Sammaltunturi. The retro plume release height was set to 560-660 m ASL, as the terrain height in ERA5 at the site was approximately 300 m ASL. The PES output resolution was set to 0.2° latitude and longitude with a 250-m height resolution up to 5 km and two additional output levels at 10 km and 50 km. “

RC7: “l. 244: *Subtitle 3.1 should be bold as 3.2 and 3.3.*”

AA7: The above suggestion was accepted

RC8: “l. 245: *The main message of the figure is not mentioned and should include something like: It shows the seasonal range of temperatures from on average XX°C in September to -XX°C in November and its interannual variability.*”

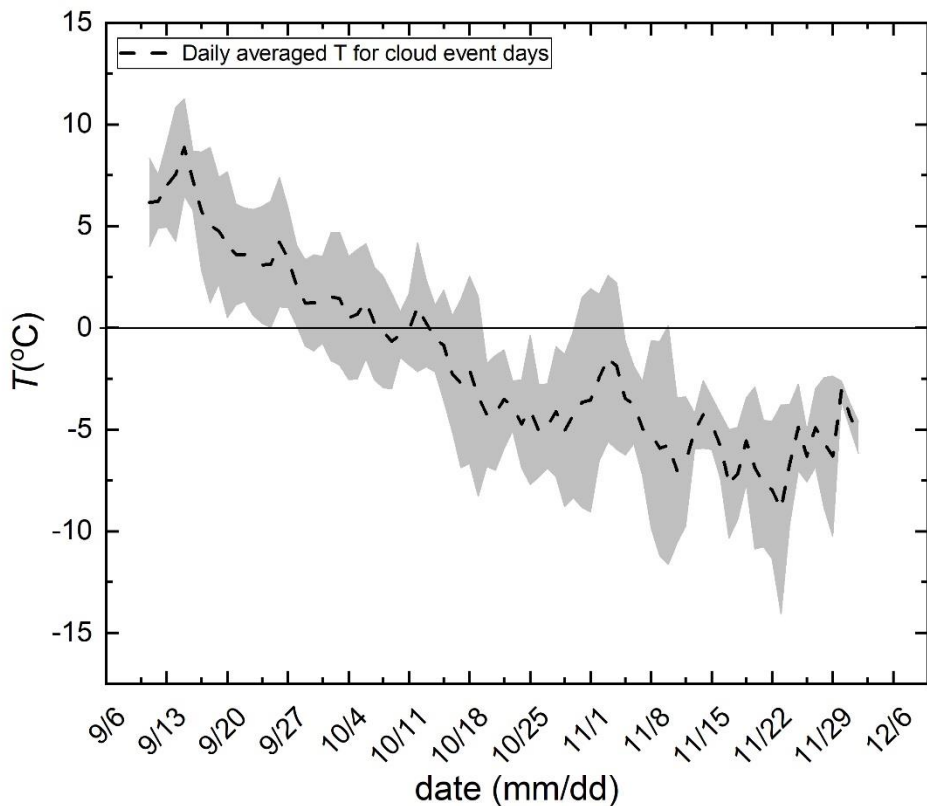
AA8: The above suggestion was accepted. The following text was added in the revised manuscript.

Line 253“ ..events”. The seasonal range of temperatures from on average 4.5 °C (SD 2.1°C) in September to -5.3 °C (SD 1.8°C) in November and its interannual variability is revealed. Days..”

RC9: “*Fig. 5 (also Fig. 7): Is there a reason to present each year separately? If not I suggest in accordance with the main message of the figure to present only one average line together with the standard deviation and include data gaps in the data section. This also applies to Fig. 7 and would increase clarity of the figures as 4 panels can even be summarized in one panel (4 lines - 4 regions). Data gaps and instrument specifications can be moved to the data section.*”

AA9:

Both figures 5 and 7 were simplified as suggested by reviewer. One average line from all PaCEs was used in the figure of the revised manuscript. Thus, figure 5 was modified as



Manuscript figure 5: The daily averaged temperatures at the Sammaltunturi site for days with cloud events during all PaCE campaigns. The black solid line is used as a reference line for 0 °C temperature. The definition of a cloud event is provided in the text. The shaded area represents the corresponding standard deviations.

Combining the data would be ideal for understanding the size distribution that corresponds to each region. To simplify our results a new figure will be provided including the average size distribution from both instruments that corresponds to each region. The instruments are still presented separately since they have different bin sizes, thus they cannot be combined. Also, they represent different measurement periods as both spectrometers were not always working at the same time. Figure 7 (Figure 8 in the revised manuscript) was modified as

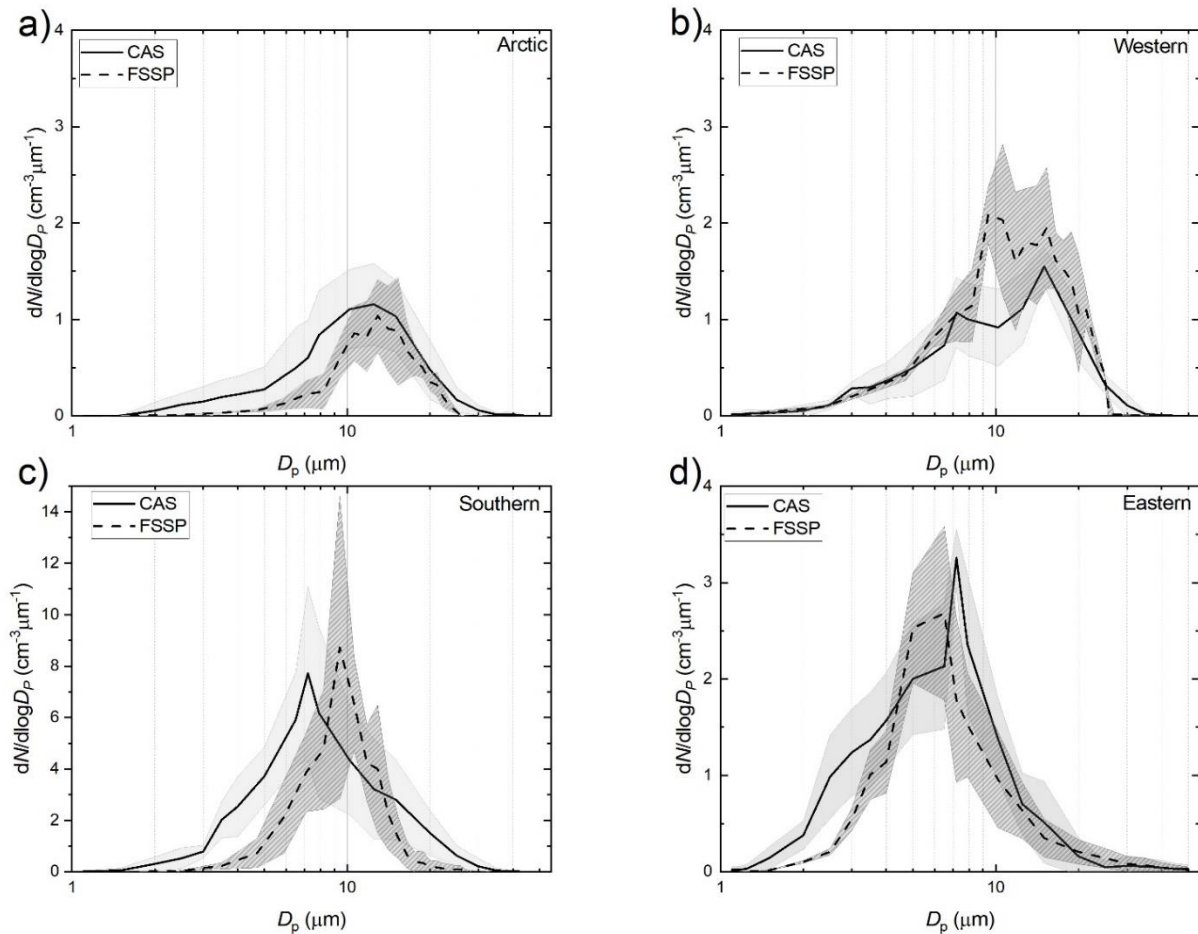


Figure 8. Cloud droplet size distribution associated with the (a) Arctic, (b) Western, (c) Southern and (d) Eastern region as they were measured by the cloud and aerosol spectrometer (CAS) and the forward-scattering spectrometer probe (FSSP) during all PaCEs. The shaded areas represent the corresponding standard deviations.

RC10: " l. 271: anthropogenic aerosols: Is this an assumption, provide a reference?"

AA10: Text was modified, and the following reference will be added.

Line 285 "... a marine environment that the natural Arctic background conditions are significantly increasing (Pernov et al., 2022))"

RC11: l. 275: Fig. 6 a) What is the meaning of the cyan color? If not necessary please remove it. If it is representing a range, please indicate it in the legend.

AA11: The shaded area represents the corresponding standard deviations. In the revised manuscript the use of shaded areas is explained.

RC12: l. 275: Fig. 6 b) Symbols (stars and circles) representing different Nc measurements are difficult to distinguish. Would recommend either summarizing campaigns sorted by PES and



CAS/FSSP (4 symbols per air mass) or summarizing it even further only by PES. If this is no option, increasing maker size and distance between campaigns would improve clarity.

AA12: The decision to present each PaCE was made due to each campaign had different operation times and the instruments could be also operative in different periods. We would like to keep each campaign and instrument to demonstrate that there were no obvious changes through years or possible malfunction of the instruments that were used and could produce biased results. However, the clarity of this figure should be improved. Thus, figure 6b was replaced and includes additional information to distinguish the years. A comment was added in the revised manuscript to note that there is no indication of dependence of the  $N_c$  through different years of measurements.

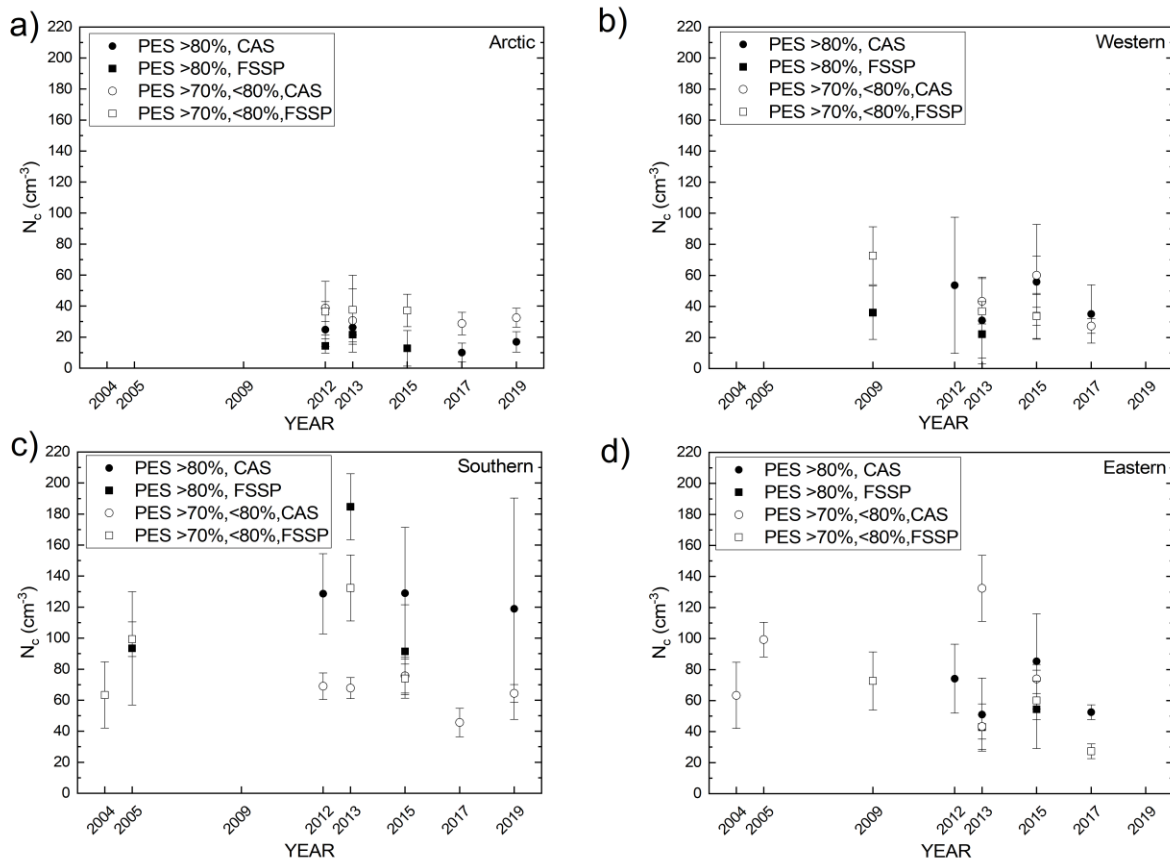


Figure 7: Cloud droplet number concentration ( $N_c$ ) for each region and single PaCE campaign as they were measured by the cloud and aerosol spectrometer (CAS) and the forward-scattering spectrometer probe (FSSP) where the PES fraction was within one region >80 % and the PES fraction was within one region from 70 to 80 %. Error bars indicate the corresponding standard deviation.

Line 308 “We present each campaign and instrument to demonstrate that there were no obvious changes through years or possible malfunction of the instruments that were used and could produce biased results”.

Line 323“...respectively. Furthermore, there was no clear indication that there was any trend in  $N_c$  through different years of PaCEs.”

RC13: l. 284 CAPS --> CAS? (as in the legend of 6b), please check usage throughout the manuscript.

AA13: In this work, only the CAS probe was used from the CAPS probe ground setup. Thus, CAS will be used in whole manuscript.

RC14: l. 395-396 Fixed vertical position, but different layers? Something is missing: "...cases that we sampled WITH different layers".

AA14: The typo was corrected, "with" was added.

RC15: l. 409 Fig 10 a and b. I cannot see the difference between CAS and FSSP in the plot. If the difference is not important for conveying the message that MVD/ED is not dependent on the position of the probe I would skip the legend entry. This figure could be improved by using a scatter density plot (2-D histogram) and regression line.

*Further, If there is no dependence between MVD/ED and position of the probe, is this something still relevant for the main message of the paper and would it require a figure plus subsection? If the answer is no I recommend skipping it or putting it in the supplementary.*

AA15: There are several difficulties to conduct in situ cloud measurements. The reason behind our choice to include this discussion as a subsection even though there is no clear dependence between MVD/ED and the position of the probe is to make it clear to the reader that we took into consideration the uncertainties that could be produced from the relative vertical position of the probe with respect to the cloud base altitude. Naturally, some microphysical properties of a cloud are dependent in the air mass origin, while some are determined by the temperature at cloud base and the height of the measurement above cloud base, and some due to the amount of vertical motion and entrainment within the vertical profile. Theoretically, the size parameters ED and MVD are expected to show a dependence on the vertical position of the probe with respect to the cloud base altitude, but they are also dependent on the temperature and initial cloud droplet number concentration at cloud base (Brenguier, 1991). Hence, we wish to show that, while cloud droplet number concentration can clearly be linked to air mass origin, it is more challenging to directly link air mass origin and size parameters (e.g. for comparison with satellite retrievals) without including the cloud microphysical processes happening in the vertical profile. We agree that the clarity of the figure should be improved, and that, in this case, it is not crucial to distinguish between the two instruments. Thus, we created a figure that presents a statistical description of MVD in 5 different altitudes above cloud base. We can see that there was no strong dependency between the vertical position of CAS and FSSP in the cloud and MVD.

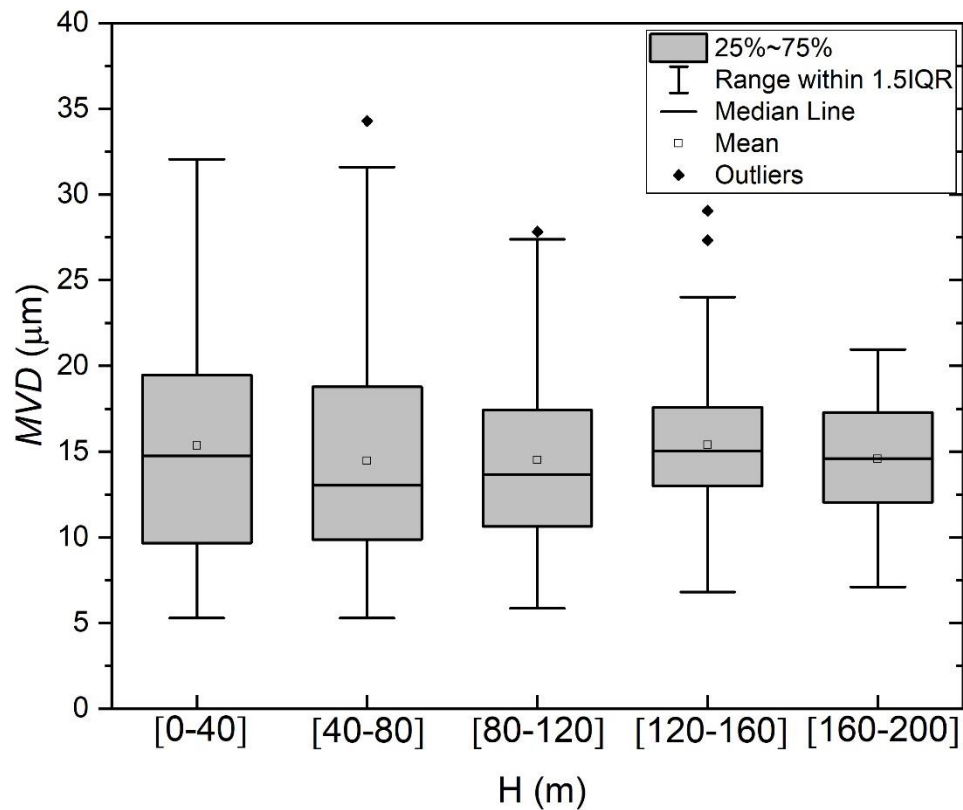


Figure 10: Statistical description of hourly averages of median volume diameter ( $MVD$ ) as they were measured by the cloud and aerosol spectrometer (CAS) and the forward-scattering spectrometer probe (FSSP) where PES was within one region  $>80\%$  for five different levels of the position of the probes inside the cloud ( $H$ ) (relative distance of the cloud ground-based spectrometer). Cloud base was measured at the Kenttäröva station.

RC16: l. 428: *to be representative or considered as representative*

AA16: The above typo was corrected “considered as representative”

RC17: l. 429: *Why are clouds more frequent when air masses originate from Southern and Eastern regions?*

AA17:

It is expected that clouds occurrence depends on the different meteorological conditions that were associated with the different air parcels. The text was modified to answer the reviewer comment.

Line 471 “..regions. This result suggests that clouds occurrence depended on the different meteorological conditions that were associated with the different air parcels. Continental ..”

RC18:” l. 440: *What kind of measurements are needed?*”

AA18: We agree with the reviewer that we should elaborate our thoughts. We highlight the need for a bigger amount of cloud measurements that will cover and allow us to investigate a wider temperature range. Particularly, we need to obtain all year around measurements to investigate the temperature dependence. The text was modified.

Instead of “However, more measurements are needed to confirm such temperature dependency of droplet sizes”

To line 494” All year round in situ cloud measurements in the area are of high importance to confirm such temperature dependency of droplet sizes. A larger data set containing a wider temperature range needs to be obtained.”

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