

We sincerely thank reviewer #2 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 as reviewer comment (RC) and authors answer (AA).

In their study Doulgeris et al combine a long-term dataset of in-situ observed cloud microphysical properties at a sub-arctic location with simulations of air mass transport. While the general methodology, uniqueness of the dataset and presentation are reasonable for publication, I do share the concerns of the first referee regarding scientific relevance and quality. In my opinion the manuscript should be reconsidered after major revisions.

General comments

RC1: *"The authors should point out more clearly where their work extends the current level of scientific knowledge. As the authors describe in the literature overview, the Twomey effect is well confirmed, and no significant additions are provided in the manuscript. It should be considered to change the manuscript type and focus to a measurement report instead of a research article."*

AA1:

The question that is raised is similar as stated by the reviewer 1, accordingly the answers are similar too. We have modified the text to be clearer where this work extends the current level of scientific knowledge. Even though there is an increased demand for long term continuous ground based in-situ cloud measurements, unfortunately there is limited instrumentation available 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 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. To our knowledge, this is the first study that connects extensive in situ cloud measurements to air mass origin. 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 Twomey effect was confirmed however in this work we mainly investigate cloud microphysics and their connection to air mass origin. We point out that there is need of considering not only local meteorological parameters but also the air mass origin in investigations of cloud processes. PaCE measuring period (during autumn) is crucial as it is a unique opportunity to get Arctic pristine air masses (Pernov et al., 2022) and combine them with in situ cloud measurements. Moreover, the procedure of distinguishing cases that correspond to one air mass origin and not to mixed one is complicated and require 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 one air mass origin). 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. As a result, we consider this work not just as a measurement report but as a research article that investigating the connection of several microphysical parameters to the cloud origin.

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°C (SD 2.5°C), -2.2°C (SD 5.9°C), 1.3°C (SD 3.9°C) and -2.8°C (SD 2.01°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”

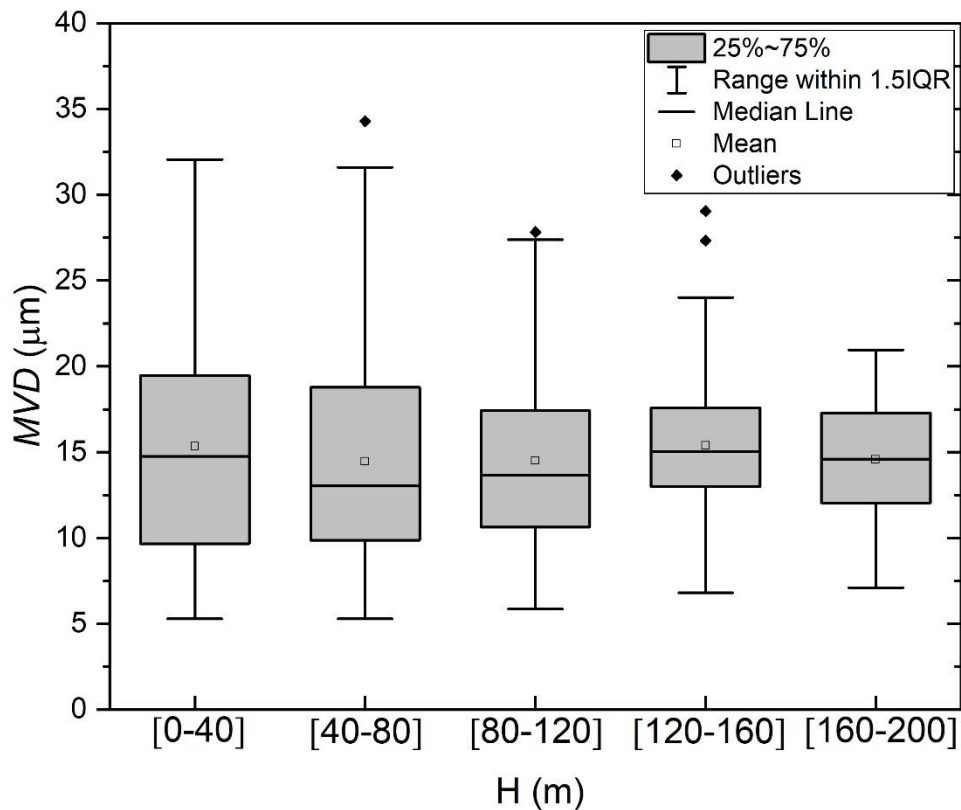
“The lowest values of cloud droplet concentration were related to clean arctic airmasses. We observed a clear relationship between air mass origin and cloud droplet number concentration. According to theoretical considerations, (Brenguier 1991; Pawlowska et al., 2006) the measurements of cloud droplet number concentration does 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:” It does not become clear how including the cloud base height information in Sec. 3.4 supports the manuscript. A distance of 4km of the ceilometer for cloud-base height retrieval seems quite far away. Also, it does not become clear if and how only stratiform cases are selected. The resulting Fig 10 looks more like a ‘point cloud’ without the chance to identify any physical relationship.”

AA2:

The cloud base height information is used to determine whether cloud events are stratiform or not. The altitude difference between the mountain top station and ceilometer station (210 m) ensures that fog cases are not selected (the liquid cloud base should be above the ground at the ceilometer location) and cloud events must be present at both locations at the same time for a minimum duration of 30 minutes and not varying much in height with time to ensure that the cloud field is stratiform rather than cumulus. The clarity of figure 10 was improved. In the new version, we present a statistical description of MVD in 5 different altitude levels of the cloud. We can see that there was no strong dependency between the vertical position of CAS and FSSP in the cloud and the MVD.



Manuscript 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.

RC3: *“The air mass source analysis raises some questions as well. The regions seem rather inconsistent. E.g., why is the Kola peninsula ‘Eastern’ and not ‘Arctic’ or why is Scotland an Ireland ‘Western’ while England is ‘Southern’. The simulation duration of 4 days is quite short. Was there any sensitivity analysis performed with 7- or 10-day simulations? How were contributions from outside the area of Fig. 3 treated?”*

AA3:

We see this work as a continuation of previous studies that were conducted at Sammaltunturi (e.g., Aalto et.al., 2003, Asmi et al., 2011). For this reason, we decided to adopt the same source areas, although the borders between the different regions are drawn on a rather coarse scale. Detailed borders of the source regions are given in Table 1 below and included in supplementary information of the revised manuscript; these criteria were used outside the area of Fig. 3. 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).

Including Kola peninsula in the Eastern rather than Arctic region ensures that the substantial anthropogenic emissions sources there (e.g., Kyrö et al., 2014) do not mask the remote Arctic air characteristics. As to the Western sector, Ireland, and Scotland (as well as Iceland and Greenland) could be excluded if the areas were redefined. However, the analysis in this manuscript indicates that small contributions (up to 20% of PES, Fig. 6 in manuscript) from other source areas is not critical for the results interpretation. Therefore, we expect that small changes to the borders of the regions would have only a very minor effect on the presented results.

In this analysis, we consider that the transport during the previous 96h is sufficient to classify the air masses into the relatively broad categories. We consider that the requirement of >80% PES within one region during the 96h is a strict criterion. Also, four days period is quite commonly used duration in air mass history analysis for ground-based in-situ measurements (e.g., Asmi et al., 2011; Makonnen et al, 2012, Riuttanen et al., 2013), as aerosols are relatively short-lived in boundary layer. In some cases, such as within the arctic during the polar night or if e.g., long-range transported forest fire smoke is present, longer simulations would be beneficial. However, this is not the case at Pallas during the measurements used here. Therefore, we have not carried out sensitivity analysis with 7- or 10-day simulations but expect that a large portion of the longer air mass history would lie outside of the area in Fig. 3 (see also Fig. 4 example case).

Table 1. Latitude and longitude ranges for each sector.

Sectors	Latitude (x)	Longitude (y)
Arctic, marine, area I	$x \geq 70^\circ \text{ N}$	

Eastern, continental, area II	$x < 70^\circ \text{ N}$	$y > 30^\circ \text{ E}$
Southern, continental, area III	$x < 65^\circ \text{ N}$	$10 < y < 30^\circ \text{ E}$
	$x < 63^\circ \text{ N}$	$5 < y < 10^\circ \text{ E}$
	$x < 55^\circ \text{ N}$	$5^\circ \text{ W} \leq y < 5^\circ \text{ E}$
Western, marine, area IV	$65 \leq x \leq 70^\circ \text{ N}$	$10 < y < 15^\circ \text{ E}$
	$63 \leq x \leq 70^\circ \text{ N}$	$5 < y < 10^\circ \text{ E}$
	$55 \leq x \leq 70^\circ \text{ N}$	$5^\circ \text{ W} \leq y \leq 5^\circ \text{ E}$
	$x \leq 70^\circ \text{ N}$	$y \leq 5^\circ \text{ W}$
Local, continental, area V	$65 < x < 70^\circ \text{ N}$	$15 < y < 30^\circ \text{ E}$

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

RC4:” L31: The statement on larger droplets in warm clouds in the current form is not supported by the presented data. Fig 8 b, d shows a decrease of particle size for the ‘Arctic’ subsample in the FSSP data.”

AA4: In majority of the cases during PaCEs, cloud droplets appeared to be more prone to grow at temperatures larger than -2° C , however it is true that in Fig 8 b, d there is a decrease of particle size for the ‘Arctic’ subsample in the FSSP data. This is due to the different amount of data in each temperature bin. For the Arctic region, the observation hours in the last bin were smaller in comparison with the other temperature bins. An explanation was added in the revised manuscript. Also, the number of samples per bin was added in the Supplementary Materials (SM) of the manuscript.

Line 384 “..spectrum. The decrease of particle size for the ‘Arctic’ subsample in the FSSP data above 0° C was due to the relatively low amount of observation in this temperature range (2 hours of observation). The observation hours related to each temperature bin for each PaCE are presented in Table 3 of the SM.”

Table S3. Observation hours related to temperature bin and each region for all PaCEs.

Temperature bin ($^{\circ}\text{C}$)	Arctic(h)	Eastern(h)	Southern(h)	Western(h)
(-10,-6)	32	99	0	0
(-6,-2)	39	85	52	45
(-2,2)	45	39	49	59
(2,6)	2	52	51	14
TOTAL	118	275	152	118

RC5: “L44: The issue of varying meteorological conditions is raised, but throughout the manuscript it does not become clear how different temperature and humidity within an air mass origin category are treated (or if they are uniform enough to be disregarded).”

AA5: Average temperatures per region as measured in situ are provided below and were added in the manuscript (line 318). In the presented dataset, temperature range per region is not wide enough to notice crucial changes in the microphysical properties of the cloud and air mass origin seem to be the most crucial parameter. N_c was not strongly dependent on temperature in this dataset. MVD and ED had minor changes (less than $1\mu\text{m}$) in several cases, e.g., eastern region from -8 to 0°C . The dependence of ED and MD on temperature was discussed in section 3.3. Relative humidity values measured at the station during cloud event were always approximately 100 %. Meteorological parameters at different altitudes at the Sammaltunturi station were also analyzed using ERA 5 re analysis and the results are provided to SM. Based on ERA5 profiles T and Q variability is smallest in Arctic air mass. The Eastern and Western air mass medians are close to the Arctic air mass, but the Southern sector has clearly higher T and Q.

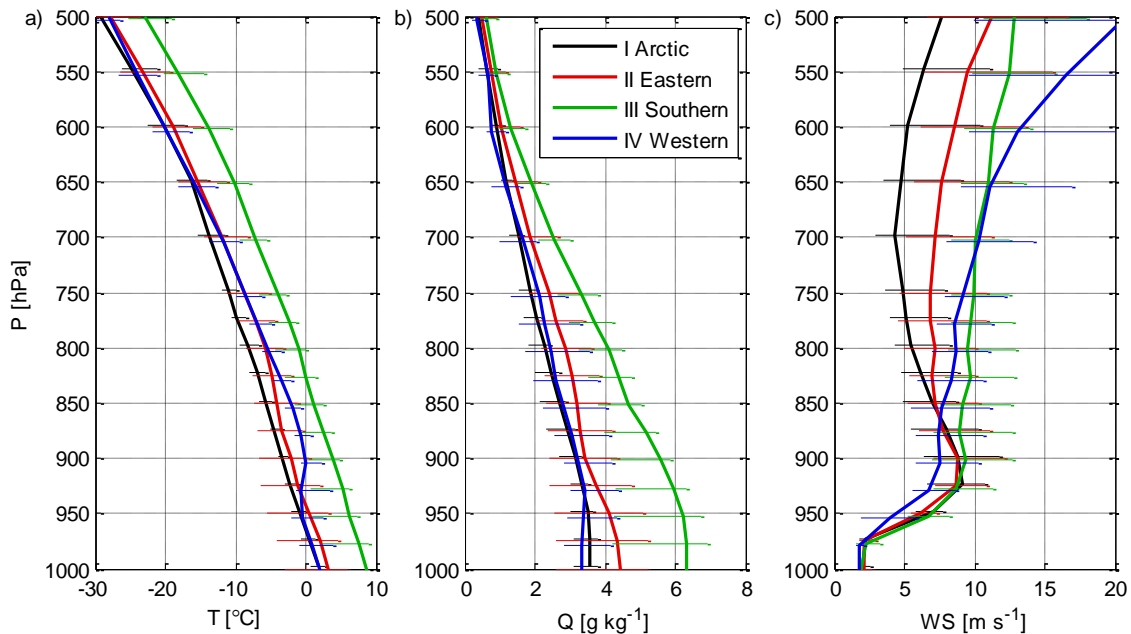


Figure S1. ERA5 temperature(T), specific humidity(Q) and wind speed (WS)profiles for the cases, when at least 80% of PES was within a source region. Line is the median and error bars indicate upper and lower quartiles. I, II, III and IV corresponds to the arctic, eastern, southern, and western region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

Line 321 “Averaged temperatures at Sammaltunturi for each air mass were -3.1°C (SD 2.5°C), -2.2°C (SD 5.9°C), 1.3°C (SD 3.9°C) and -2.8°C (SD 2.01°C) for the arctic, eastern, southern and western region respectively.”

RC6: “L49-51: Consider rephrasing this sentence, it is hard to grasp what is reason of the limited knowledge and what is the consequence.”

AA6: Text was modified as the reviewer suggested.

Line 52 “It is important to understand how different air masses can influence the aerosols and the cloud microphysics when the cloud dynamics and the interaction between aerosols and clouds are examined (e.g., Painemal et al., 2014; Orbe et al., 2015a; Fuchs et al., 2017; Cho et al., 2021).”

RC7: “ L79-83: The sudden appearance of ice particle sizes confuses the reader. As the manuscript focuses only on cloud droplets, consider removing it”

AA7: The introduction is focusing on clouds and air mass origin. This work is one the few studies that investigate long term cloud properties so we would like to keep this reference in our introduction. However, as the reviewer suggested we will modify the text to avoid any readers’ confusion.

RC8: *“Fig 1: The information content of this map has to be increased. Include the elevation, ideally as shading or contour line, as you later argue based on the orography. A legend and lat-lon grid are lacking. Does the darker green color indicate forest? The labels are too small.”*

AA8: The above suggestion was accepted; A new map was created.

RC9: *“L147 and 180: Please provide a histogram of wind direction/wind strength. Are certain air mass origin categories subsampled due to filtering periods when FSSP and CAPS did not look into the same direction.”*

AA9: There were not any data subsampled in the period when FSSP and CAS did not look into the same direction. As we highlight in line 181 “We only used measurements when the cloud spectrometers were facing the wind direction”.. This is the main reason that throughout the manuscript we present the two instruments separately. The obtained data set from the two instrument setups is different due to their different operational times (see table 2). The histogram of the wind direction for the CAS probe will not provide any further information as we used data when the instrument was looking to the wind direction (225 +- 25). The histogram of the wind direction for the FSSP data set is provided below. Wind speed was in all cases lower than the probe air speed of both setups (for the FSSP 6.7 m/s (SD 2.4 m/s) and for the CAS 7.1 m/s (SD 2.3 m/s)). A detailed description of all PaCEs, both ground setups, installation, limitations and the methodology that was used is documented in previous studies (Doulgeris et al., 2020, Doulgeris et al., 2022).

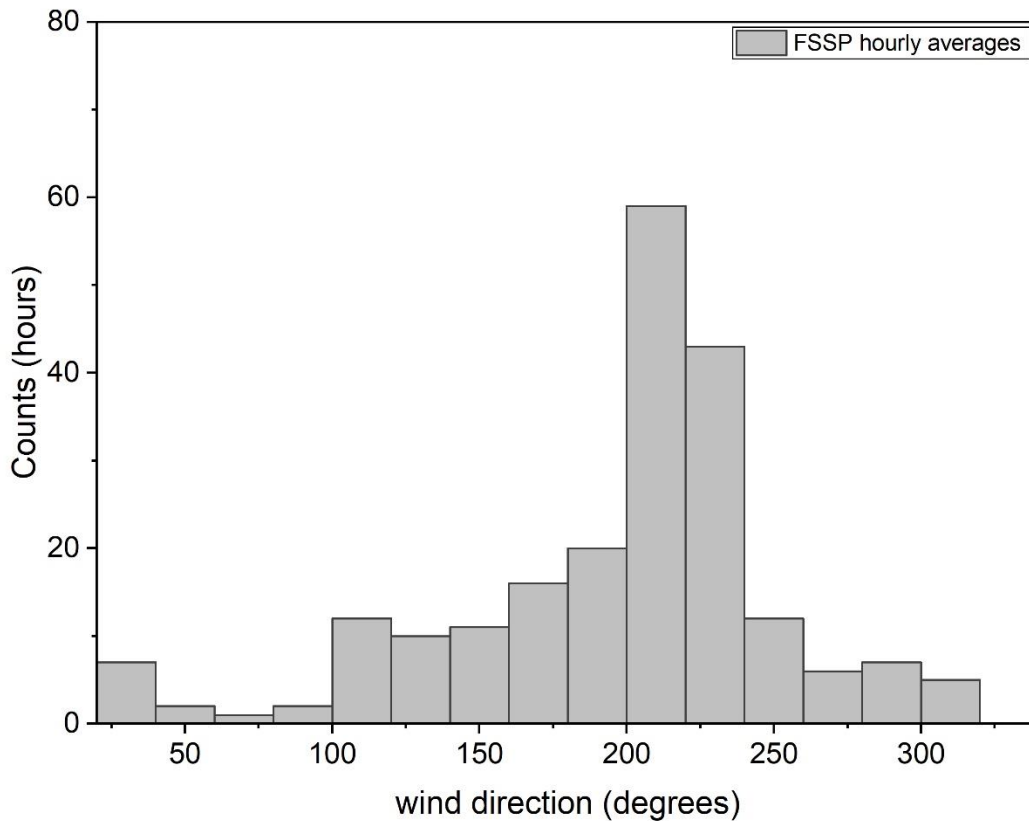


Figure 1. Wind direction histogram for the FSSP data set that was used in this work.

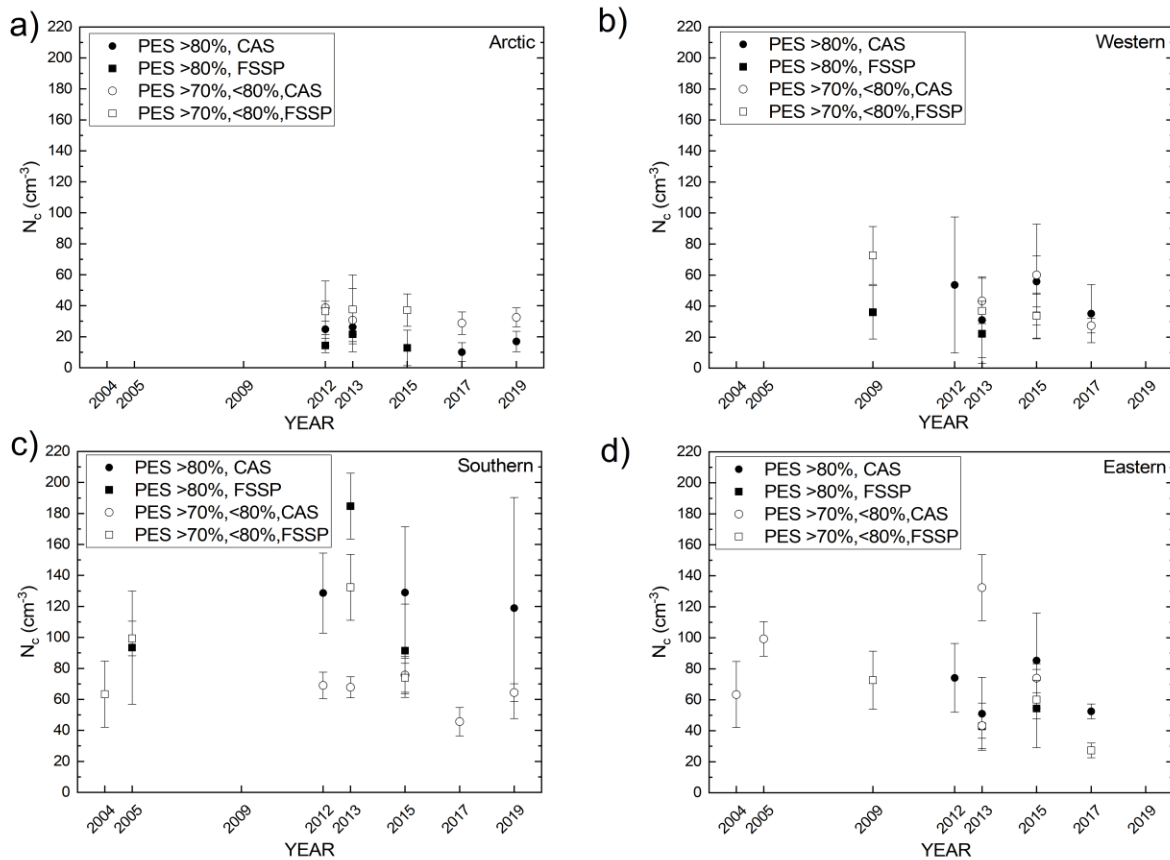
RC10: " L152-157: At which of the sites were the meteorological observations conducted? Sammaltunturi? Given the amount of detail on hardware in this paragraph, it would be nice to have that the reader would not be forced to consult Doulgeris 2020/2022 for this piece of information."

AA10: Meteorological observations were conducted at the Sammaltunturi site. Text was modified as the reviewer suggested.

Line 156 "... that was deployed at the Sammaltunturi site. All.."

RC11: "Fig 6 b: Please distinguish the years. Make clearer what data is from CAS and what from FSSP"

The clarity of figure 6b was improved, years were added. Thus, fig. 5b was replaced by fig.7 of the revised manuscript.



Manuscript 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 % (full symbols) and the PES fraction was within one region from 70 to 80 % (open symbols). Error bars indicate the corresponding standard deviation.

RC12: “L304: Please provide the duration of the >80% periods also as fraction of the total in-cloud duration”

AA12: In total 2004 hours of cloud observations resulted in 706 hours of cloud observations that related to non-mixed air mass origins. The following information was added

Line 322: “..probe (from total 2004 hours of in situ cloud data 706 hours belongs to non-mixed air mas origin.)”

RC13: ”Fig 7: Without indicating the variability, the yearly averages are of limited use for the reader. How may hours of observations are available for each year and each cluster? Also, given the typical duration of the campaigns, shown is an autumn average.”

AA13: Variability information were added in the Supplementary Materials (SM) of the revised manuscript, for each year and region. Figure 7 was updated and simplified. We also agree that “yearly average” is misleading the reader, and it was changed to PaCEs average.

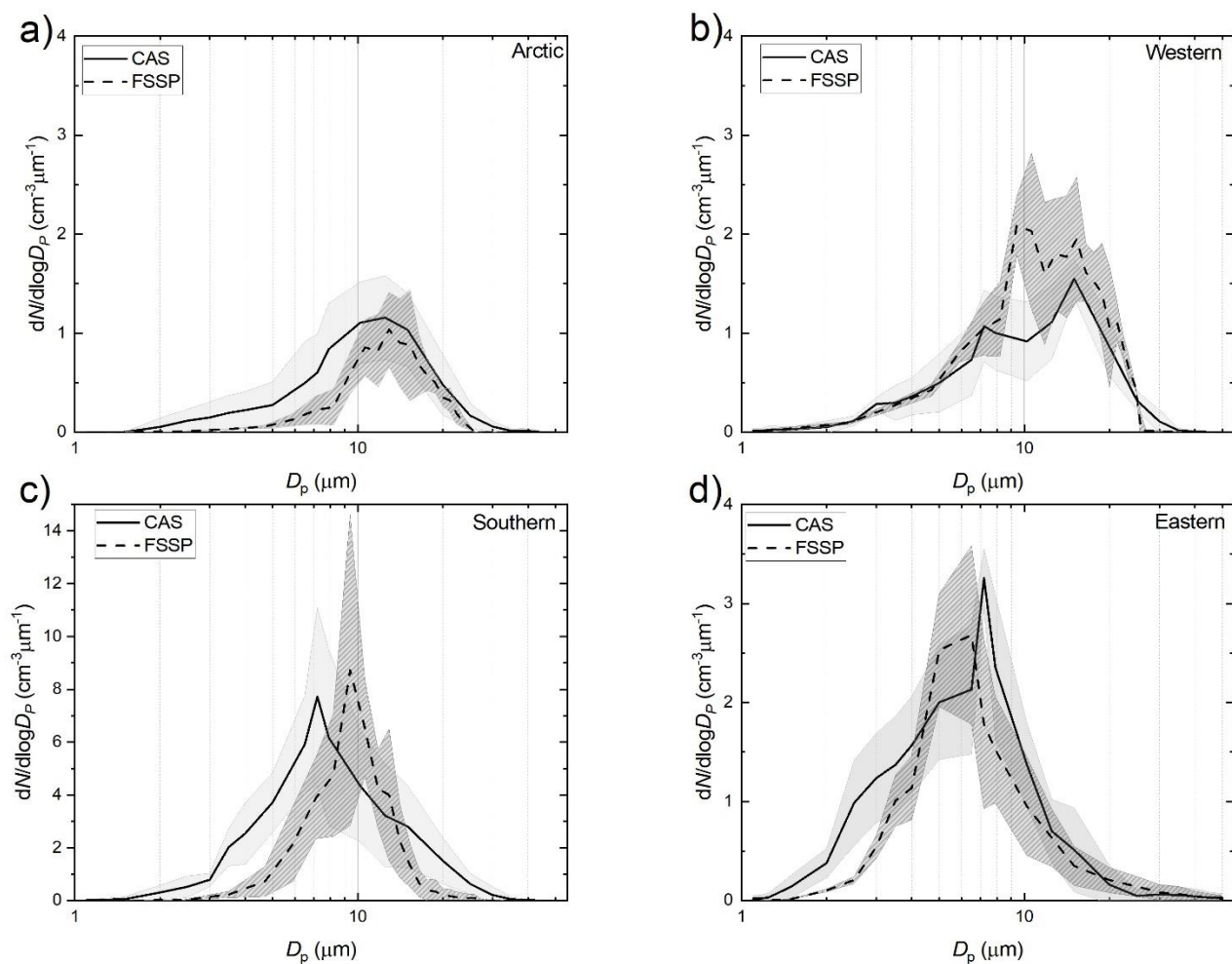


Figure 8. Cloud droplet size distribution associated with the (a) Arctic, (b) Western, (c) Southern and (d) Eastern region as they were measured by the cloud and aerosol spectrometer (CAS) and the forward-scattering spectrometer probe (FSSP) during all PaCEs.

Table S2. In total 706 observation hours of non-mixed airmasses related to each region for each PaCE.

	Arctic(h)	Eastern(h)	Southern(h)	Western(h)
2005	0	0	11	0
2009	0	0	29	9
2012	30	53	0	10
2013	22	54	16	25
2015	8	138	58	46
2017	18	30	0	28
2019	40	0	38	0
TOTAL	118	275	152	118

RC14: “L350-352: The statement on shorter lifetime of warm Arctic clouds not well supported by the data presented. Please either extend the argumentation or omit that sentence.”

AA14: Sentence was omitted as the reviewer suggested.

RC15: “Fig 8: Please include the no of samples per bin, instead the vague statement in L354.”

AA15: The number of samples per bin was added in the SM of revised manuscript.

Table S3. Observation hours related to temperature bin and each region for all PaCEs.

Temperature bin ($^{\circ}\text{C}$)	Arctic(h)	Eastern(h)	Southern(h)	Western(h)
(-10,-6)	32	99	0	0
(-6,-2)	39	85	52	45
(-2,2)	45	39	49	59
(2,6)	2	52	51	14
TOTAL	118	275	152	118

RC16: “L367: How did you account for different temperatures in different air masses for this conclusion?”

AA16: This finding is indeed attributed to the typical temperatures at cloud base for each air mass origin, as the temperature at cloud base determines the gradient of LWC with height (Brenguier 1991). Temperatures at cloud base were not directly obtained but the temperature at the Sammaltunturi measurement station provides a good estimation. Air masses with colder temperatures showed lower LWC values, and the air mass with the broadest temperature range also showed a wider range of LWC values.

A sentence has been added earlier in the manuscript at Line 318: “Averaged temperatures at Sammaltunturi for each air mass were -3.1°C (SD 2.5°C), -2.2°C (SD 5.9°C), 1.3°C (SD 3.9°C) and -2.8°C (SD 2.01°C) for the arctic, eastern southern and western region respectively.”

RC17: Fig 9 a: similarly to Fig 6b, please indicate the single years and make CAPS and FSSP more distinguishable

AA17: In the revised manuscript after updating Fig. 7 (or previously 6b) we show that there is not a clear yearly dependence in N_c . Thus, we proved that there were no obvious changes through years or possible malfunction of the instruments that were used and could produce biased results. So, we decided that the best option would be to simplify Figure 9a (figure 10a in latest version) and include only the statistical description.

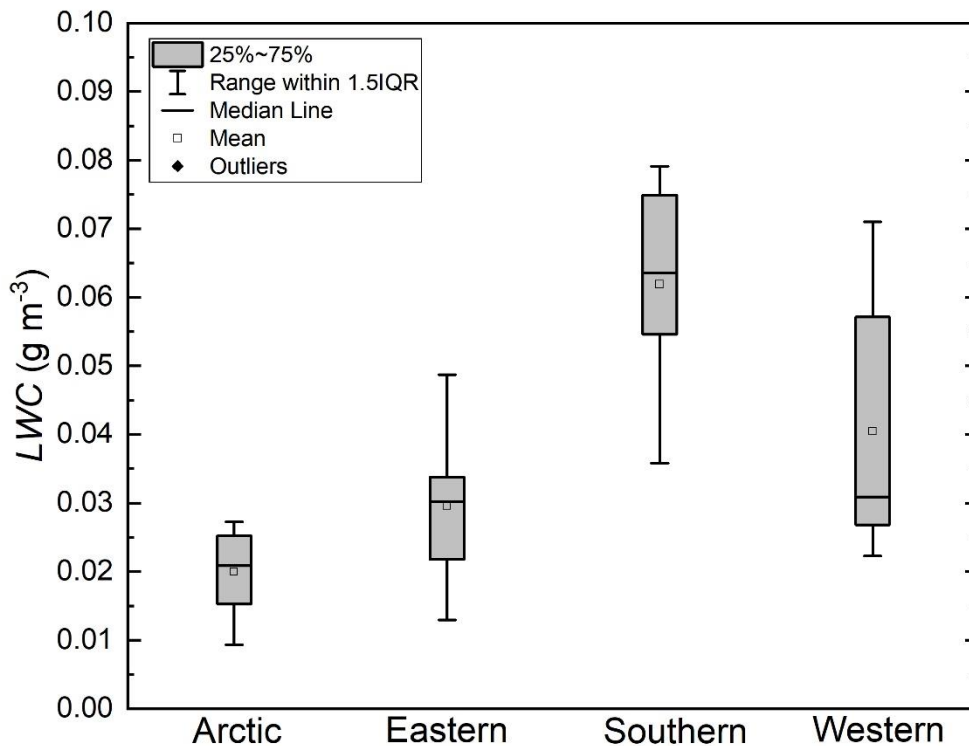


Figure 9: Statistical description of liquid water content (LWC) for each region as it was measured by the cloud and aerosol spectrometer (CAS) and the forward-scattering spectrometer probe (FSSP) where PES was within one region >80 %.

RC18: “L431-434: The conclusions on number concentration and diameter with respect to the height are not backed by the analysis presented in Sec 3.4 at all. Either remove this aspect or expand on the reasoning, including descriptive figures.”

AA18: We have rewritten this statement. What we wanted to highlight in the conclusion is that, according to theory, the measurement of cloud droplet number concentration should not depend on the height into cloud at which the measurement is made (Pawlowska et al., 2006), therefore relationships between air mass origin and cloud droplet number concentration should be possible to observe. Such relationships are not expected between air mass origin and droplet size (Brenquier 1991; Boers et al., 2000) as these require additional considerations (temperature at cloud base, cloud droplet number concentration, rate of increase of LWC with height, height of measurement above cloud base).

Replace: “Number concentration was expected to be a robust signal with no dependency from the vertical position of cloud spectrometer in the cloud. On the other hand, both effective radius and median volume diameter has some extra uncertainties depending on the altitude with respect to cloud base.”

with line 482 “We observed a clear relationship between air mass origin and cloud droplet number concentration. According to theoretical considerations (Brenguier 1991; Pawlowska et al., 2006), the measurements of cloud droplet number concentration does not depend on the vertical position of the cloud spectrometer within the cloud layer.”

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