Response to Reviewer Comments: Manuscript "Frequent occurrence of newly formed aerosol particles over wide geographical areas in the Arctic free troposphere and atmospheric boundary layer"

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Reviewer #1: The authors present rare airborne measurements of aerosol and meteorological parameters over the Arctic ocean. Quantifying the relative role of the various sources of Arctic cloud condensation nuclei (CCN) is a challenging task, but necessary in order to improve model uncertainty. Overall, the paper is well-written and I think the results of this study are a valuable contribution to the field as they provide insight into the potential role of new particle formation over the sea ice surface as well as aloft.
However, I believe more transparency is needed regarding the methodology of the study in order for the results to be interpreted correctly by the reader. My suggestion is to consider this manuscript for publication after major revisions, as I believe significant changes need to be made. Most of the changes involve including analysis into the manuscript that I am pretty sure the authors have already conducted, but not shown, so the work needed is probably not extensive. I also think that there are some claims that need to be toned down, rephrased, or further contextualized, as the results of this study are not enough to support them.

Author reply: We thank the reviewer very much for the comments and constructive criticism to our manuscript. We have carefully revised the manuscript based on the listed points.
Please find our detailed response below. The reviewer's comments are marked in black, our respective answers in green, and resulting changes in the manuscript and supporting information in blue.

General comments

i. The entire analysis leans on the identification method of newly formed particles, which is nicely demonstrated and explained - in the SI. I absolutely believe that this explanation and exemplary figure are essential parts of the methods and thus belong in the actual manuscript, to convince the reader you have in fact correctly identified particles sourced by new particle formation (NPF).

We thank the reviewer very much for this suggestion. Motivated by this comment, we have moved the figure and corresponding discussion to the methods section of the main manuscript. The respective paragraph in Sect. 2.2 now reads (lines 99 to 108 in the revised manuscript):
To detect newly formed particles, we fitted mono-modal, bi-modal, or tri-modal log-normal functions to the MPSS-measured PNSDs, with the different modes representing nucleation, Aitken, and accumulation modes, respectively. Following Kulmala et al. (2004), we refer to aerosol particles in the measured dry diameter range from below 10 nm to 20 nm as nucleation mode particles, from 20 nm to 90 nm as Aitken mode particles, and from 90 nm to 600 nm as accumulation mode particles. We consider an MPSS-measured PNSD as indicative for newly formed particles if the fitted distribution features a nucleation mode with a geometric mean diameter below 10 nm, i.e., a mode with a maximum below the lower size limit of the MPSS. The detection of newly formed particles was verified by visual inspection of the fitted PNSDs. Only distributions measured at a constant altitude were considered for quantitative analysis, i.e., PNSDs measured during the aircraft's ascents and descents were not taken into account in quantitative terms. Fig. 1 shows examples of three fitted PNSDs, with the data displayed in Fig. 1a and Fig. 1c indicating the presence of newly formed particles, and the data in Fig. 1b the absence of newly formed particles.

ii. The statistics of the observations are only poorly described, leaving an important lack of context for the reader and the potential risk for miss- and overinterpretation. In section 3.1 I suggest adding how much flight time was spent in the boundary layer (BL) and free troposphere (FT) respectively, at what frequency newly formed particles (NFP) were identified in the BL (regardless of surface type) and FT, and then the relative frequency of NFP over sea ice and in the vicinity of clouds. It is important to show to the reader how you identified the 3 atmospheric environments considered. Furthermore, it is stated more than once that no NFP were observed in the BL over open ocean, but this is kind of contradicted in the SI when explaining the 14 % non-assigned observations. In figure 1 it is clear NFP were observed over open ocean, was this then not in the BL? More importantly, how much time was actually flown over the open ocean inside the BL? Again, Figure 1 indicates not much. Please clarify and revise, as this is stated as one of your conclusions.

Thank you for this remark! With the measured data available, adding the requested statistics is impossible to fulfill in a truly quantitative manner. The

main reason is that we do not always have data concerning the atmospheric stratification along the flight track. In other words, we cannot always clearly distinguish the airplane's actual altitude being within our outside the atmospheric boundary layer (ABL). Similarly, due to missing cloud microphysical measurements, we cannot always determine the presence or absence of clouds in all air masses sampled. Nevertheless, we can provide a respective rough estimate utilizing ERA5 reanalysis data. However, the related results should be considered semi-quantitative only, as the reanalysis data, due to their inherent uncertainty and limited spatial resolution, are not well suited to describe the prevailing conditions at the aircraft's exact position.

Related statistics, such as relative frequencies, would be biased by the overall flight planning, e.g., the actual times spent at specific altitudes and/or in certain target regions. As we noted in the methods section 2.1, "our sampling is potentially biased towards viable flight conditions, e.g., high visibility at Longyearbyen Airport. Furthermore, we mostly preferred cloud-free conditions in the target regions or elevated cloud layers (cloud base $\gtrapprox 200\,\mathrm{m}$), and no precipitation." We would therefore like to stick to our original approach of not discussing the requested statistics in the main text, but still provide rough values as a broad overview in the SI. Please find the details in the revised section below.

The following section is placed in the SI (lines 32 to 64):

S3 Atmospheric conditions along the aircraft's flight track

Due to the lack of, e.g., drop sonde and cloud microphysical measurements, we do not have available continuous data concerning the atmospheric stratification and the occurrence of clouds along the flight tracks. In other words, we cannot always unambiguously distinguish the airplane's actual position being within our outside the atmospheric boundary layer (ABL). Furthermore, we cannot always verify the presence or absence of clouds in the aircraft's proximity. However, we can provide a very rough estimate of the total measurement times inside the (cloud-free) ABL, in the (cloud-free) free troposphere (FT), and in cloud vicinity based on reanalysis data from the fifth generation ECMWF reanalysis (ERA5) dataset. Thereto, we interpolated several ERA5 parameters such as specific cloud liquid water content ($CLWC$), specific cloud ice water content $CIWC$, and the ABL height on to the flight track of the Polar 6 aircraft. Here, we consider a flight section to have been carried out in the vicinity of clouds if the sum of $CLWC$ and $CIWC$, i.e., the total cloud water content (CWC) exceeds the threshold of $CWC > 0.01\,\mathrm{g\,kg^{-1}}$. The latter is based on in situ measurements of CWC of Arctic clouds conducted by McFarquhar and Cober (2004) in April 1998 and Moser et al. (2023) in April 2019. In both studies, values on the order of $CWC \sim 0.01\,\mathrm{g\,kg^{-1}}$ were reported for in-cloud measurements. We consider measurements to have taken place in the free troposphere (ABL) if the aircraft's flight altitude was above (below) the ERA5 ABL height. Here, the flight sections in cloud vicinity based on the threshold above are additionally excluded.

Based on the reanalysis data, for our campaign, we determine a total measurement time of about 25 hours and 10 minutes in the free troposphere, approxi-

mately 2 hours and 10 minutes in the vicinity of clouds, and about 8 hours and 10 minutes inside the ABL (Table 1). Here, 60 minutes were carried out inside the ABL over ocean with sea ice floes, 3 hours and 30 minutes inside the ABL over sea ice, and 2 hours inside the ABL over open, completely ice-free ocean. In this analysis, the research flight on 29 April was omitted since it was carried out inside the fjords close to Longyearbyen under a completely closed cloud deck. These measurements amount to a duration of 1 hour and 40 minutes. Observations of newly formed particles on this day ($\approx$ 10 minutes) are assigned to the unclassified category.

In addition, the relative frequencies of occurrence of newly formed particles for each environment are provided in Table 1. These values should be interpreted carefully and only provide a very rough estimate. For example, not for all flight sections dedicated to probe directly the cloud top or cloud base, i.e., legs which are known to have taken place in the direct vicinity of clouds, ERA5 predicted consistently non-zero values of CWC throughout the leg. Furthermore, during the case study on newly formed particles over sea ice, the ERA5-based ABL height was roughly 100 m lower than the ABL height determined from the vertical profiles of potential temperature derived from in situ measurements in the target region.

For all case studies and the assignment of measurement periods featuring newly formed particles to the four categories "free troposphere", "cloud vicinity", "ABL over sea ice", and "ABL over ocean with sea ice floes", we only consider flight sections for which the prevailing conditions could be clearly identified based on the in situ data. If newly formed particles were detected but either, e.g., the ABL height could not be determined for the target region or the the absence/presence of clouds close to the aircraft could not be verified by studying in situ video data, these observations are assigned to the unclassified cases.

Thank you as well for the question regarding the observations of newly formed particles over open ocean! Indeed, Fig.1 (Fig. 2 in the revised manuscript) shows that newly formed particles were found over open ocean. However, only during the research flight on 11 April (see Fig. S1 for a map of all flight paths), these particles were found inside the ABL (between 60 to 300 m). Here, the sea surface in the target region contained both sea ice and open water. The other cases over ocean the reviewer probably refers to were found on different days and at much higher altitudes in the free troposphere, i.e., not inside the ABL as correctly assumed by the reviewer.

Previously, the observations of newly formed particles on 11 April were assigned to the unclassified category. Motivated by the reviewer's comment, we investigated the unclassified cases further. About 40 % of those cases previously assigned to the unclassified category were found to have been carried out inside the ABL over ocean with sea ice floes on 11 April, i.e., with the surface below the aircraft featuring both open water areas and sea ice, as mentioned above. These cases are now included as a category in Fig. 3 (previously Fig. 2) as "newly formed particles inside the ABL over ocean with sea ice floes", summarized in the main text (Sect. 3.1), and discussed in detail in the SI in Sect. S5.

Table 1: Overview of total measurement time as well as duration and frequency of newly formed particle-occurrence (NFP-occurrence) for the four environments.

	FT	ABL irrespective of surface	ABL over sea ice	ABL over ocean with sea ice floes	cloud vicinity
Total measurement time (based on ERA5)	25 h 10 min	8 h 10 min	3 h 30 min	1 h	2 h 10 min
Duration of NFP-occurrence (based on in situ data)	4 h 40 min	2 h 20 min	1 h 40 min	40 min	2 h 40 min
Frequency of NFP-occurrence	19 %	29 %	48 %	67 %	(> 100 %) [1]

1: Nonphysical value, resulting from ERA5 not always predicting clouds throughout all legs which are known to have been carried out in cloud vicinity, according to the in situ observations. Considering those times based on the in situ data instead of the ERA5-based values, a frequency of about 73 % can be reported.

In this SI section, we also discuss the measurements (approximately 120 minutes in total) inside the ABL over open, completely ice-free ocean (on 11 and 25 April 2024, see Fig. S1), during which no newly formed particles were detected. The remaining unclassified cases of newly formed particle-observations are now also briefly discussed in Sect. 3.1 instead of in the SI as was previously the case. Thank you very much for this comment which has helped us to improve the manuscript. Please find the details in the revised sections below.

Revised paragraphs in Sect. 3.1 in the main text, lines 136 - 198):

[...] Analyzing all measurements featuring newly formed particles in the course of our campaign, we could identify four distinct environmental conditions (based on the in situ data, see Sect. S3; Fig. R1). These cases include newly formed particles in the free troposphere (without clouds being present, Sect. 3.2), newly formed particles at low altitudes in the ABL over sea ice (Sect. 3.3), newly formed particles in the vicinity of clouds (Sect. 3.4), and newly formed particles inside the ABL over ocean with sea ice floes, i.e., the sea surface consisted of both open water and sea ice during the latter observations (Sect. S5). No indications for the presence of newly formed particles were found within or near the ABL over open, completely ice-free ocean (Sect. S5).

Fig. R1a depicts the median size distributions of the PNSDs indicating the presence of newly formed particles, for each of the four environments. The highest particle number concentrations in the nucleation mode are found during observations of newly formed particles in the free troposphere. Inside the ABL over sea ice and in cloud vicinity, concentrations in the nucleation mode are smaller by a factor of roughly two. The smallest nucleation mode concentrations are observed inside the ABL over ocean with sea ice floes, with the concentrations being reduced by about one order of magnitude compared to the free tropospheric case. In the accumulation mode, roughly similar concentrations are observed during newly formed particle-occurrence inside the ABL over ocean with sea ice floes and cloud vicinity. Inside the ABL over sea ice, accumulation mode concentrations are slightly higher. The lowest accumulation mode concentrations are found in the free troposphere, with the mode being decreased by a factor of roughly two to four compared to the other cases.

The reader is referred to Sect. S4 and Fig. S3 regarding the related temperature and relative humidity conditions prevailing during observations of newly formed particles. We will discuss the observations for each of the four categories, which represent 92 % of all observations of newly formed particles, in more detail in Sect. 3.2, Sect. 3.3, Sect. 3.4, and Sect. S5.

About 45 % of all PNSDs featuring newly formed particles were detected in the free troposphere, corresponding to approximately 4 hours and 40 minutes of measurements (Fig. R1b, c) and distances flown of $\approx$ 1460 km. About 16 % ($\approx$ 1 hour and 40 minutes, $\approx$ 350 km) of all cases featuring newly formed particles were found in the ABL over sea ice, as well as 6 % ($\approx$ 40 minutes, $\approx$ 130 km) in the ABL over ocean with sea ice floes. Approximately 25 % ($\approx$ 2 hours and 40 minutes, $\approx$ 630 km) of all cases were observed in the vicinity of clouds (Fig. R1b, c). For approximately 8 % ($\approx$ 50 minutes, $\approx$ 200 km) of all PNSDs featuring

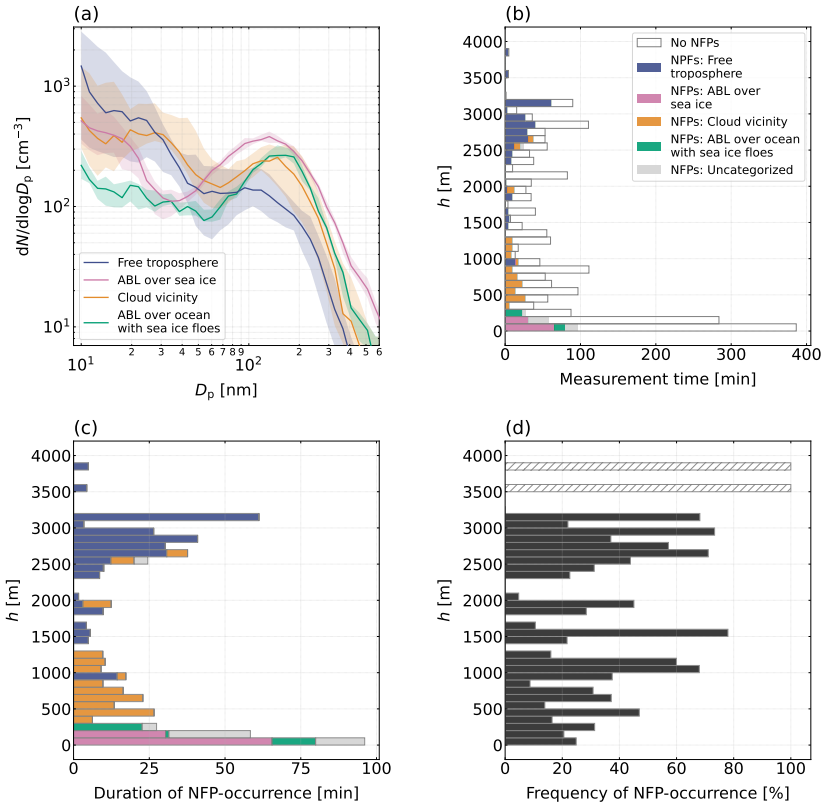

Figure R1: **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and inside the ABL over ocean with ice floes (Sect. S5). **(b)** Total measurement times per altitude (grouped into height bins with a width of 100 m) showing the absence of newly formed particles as well as times during which newly formed particles were detected for the different environments. **(c)** For convenience, this subplots shows the duration of newly formed particle-occurrence (NFP-occurrence) per measurement altitude again for the different categories, i.e., the same NFP data as in the first panel. The NFP data here and in panel (b) are shown as stacked bars. Note the different range of the x-axis compared to (b). **(d)** Relative frequency of NFP-occurrence per measurement altitude (irrespective of category), i.e., NFP-occurrence divided by total measurement time. The hatched bars at altitudes higher than 3500 m indicate that here only one full PNSD measurement was conducted within each altitude bin, while below 3200 m on average 14 MPSS scans were carried out at each altitude, with a maximum of 79 scans between 60 and 100 m (Fig. R1a).

newly formed particles, the prevailing conditions could not unambiguously be identified, i.e., these cases could not clearly be assigned to one of the categories or to another separate category (Fig. R1b, c). They include flight sections during which clouds were present, but in contrast to the newly formed particles classified as in the vicinity of clouds, we did not carry out dedicated legs directly at cloud top or base, respectively. Also included are flight legs for which it could not unequivocally be determined if the legs were conducted inside or outside the ABL (see Sect. S3).

[...]

To further elucidate the atmospheric scenarios under which newly formed particles were observed, and to gain insights into potential precursor gases and their origins, in the following sections, the results of case studies focusing on newly formed particles in the free troposphere, inside the ABL over sea ice, and in the vicinity of clouds will be presented. First, specific examples for these scenarios will be given. Second, to put the given examples into a larger perspective, we will discuss respective backward trajectory analyses for all air masses featuring newly formed particles for each of these three environments. Regarding the presence of newly formed particles inside the ABL over ocean with sea ice floes, as well as the absence of such particles inside the ABL over completely ice-free ocean, the backward trajectory analysis required is somewhat different compared to the other three cases (for example, due to the prevailing meteorological conditions, e.g., air masses passing over the island of Spitsbergen, see Sect. S5 for details). To keep the main text both consistent and concise, these latter observations are discussed comprehensively in the SI in Sect. S5.

The following sections are additionally included in the SI, discussing the measurements inside the ABL over ocean with sea ice floes and over completely ice-free ocean (lines 99 - 167):

S5 Newly formed particles inside the ABL over ocean with sea ice floes

S5.1 Meteorological conditions

During the BACSAM II campaign, we conducted two research flights which included legs inside the ABL over ocean (on 11 and 25 April, Fig. S1). On all other days, the flight sections over open ocean visible in Fig. S1 were carried out above the ABL.

On 11 April, we measured PNSDs in a target region in the lee west of Spitsbergen over ocean, with the ocean surface featuring both open water areas and patches of sea ice floes, as well as closed sea ice in the fjord in the proximity of WP7 (Fig. R2a). The air masses probed during the legs inside the ABL originated in sea ice-covered regions north and east of Svalbard (Fig. R2b, c). In the last 24 hours prior to measurements inside the ABL, easterly winds advected cold air over the snow- and ice-covered Svalbard mountains to the target region over ocean (Fig. R2b, c). While mostly clear-sky conditions prevailed, a few scattered clouds were present in the eastern parts of the target region as well (Fig. R2a).

On 25 April, measurements were carried out in a target region southwest of

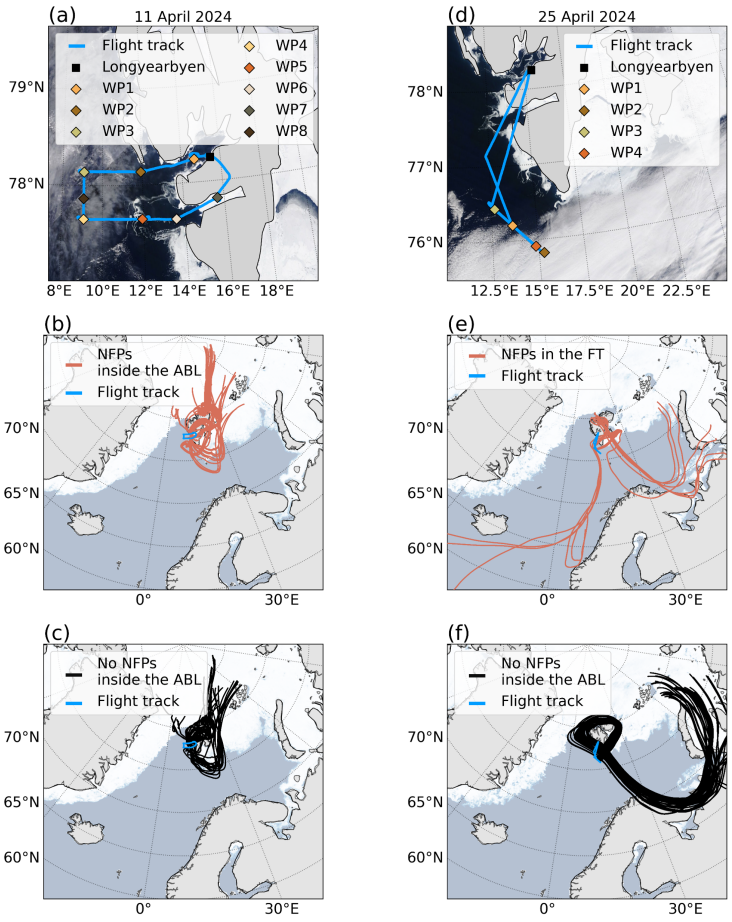

Figure R2: **(a)** Flight track of RF03 on 11 April 2024. **(b)** Five-day backward trajectories associated with newly formed particles (NFPs) inside the ABL on 11 April. **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles inside the ABL on 11 April. **(d)** Flight track of RF09 on 25 April 2024. **(e)** Five-day backward trajectories associated with newly formed particles in the free troposphere (FT) on 25 April. **(f)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles inside the ABL on 25 April. The MODIS-TERRA satellite images shown in the panels (a) and (d) were retrieved from https://wvs.earthdata.nasa.gov. The sea ice concentration data shown in the subplots (b), (c), (e) and (f) were retrieved from www.seaice.uni-bremen.de (Spreen et al. 2008).

Spitsbergen over open, completely ice-free ocean (Fig. R2d). On this day, air masses approached from southern latitudes (Fig. R2e, f). For the air masses probed inside the ABL (Fig. R2f), an anti-cyclonic flow around Svalbard resulted in north-easterly winds and advection over the sea ice and subsequently over open ocean prior to measurements inside the ABL (Fig. R2f). Air masses probed inside the ABL in the northern section of the target region additionally flowed over the Spitsbergen mountains before approaching the area over open water. In this northern section, sparse scattered cumuli clouds were present, while overcast conditions prevailed in the southern part (Fig. R2d).

S5.2 Particle number size distribution measurements

On 11 April, we carried out flight legs inside the ABL at altitudes of approximately 75 m (from about 9:10 to 9:20 UTC) and 240 m (9:25 to 9:45 UTC) in the northern section of the target region between waypoint 2 (WP2) and WP3 (left y-axis in Fig. R3a, see Fig. R2a for the location of the WPs). One leg was conducted at ≈1300 m (9:50 to 10:05 UTC) outside the ABL in the free troposphere. A similar pattern was flown in the southern section between WP4, WP5, WP6 and WP7 (left y-axis in Fig. R3a, see Fig. R2a for the location of the WPs) with legs at ≈1300 m in the free troposphere (10:25 to 10:40 UTC) and inside the ABL at ≈240 m (10:50 to 11:05 UTC), ≈75 m (11:10 to 11:40 UTC), and again at ≈240 m (11:45 to 12:00 UTC).

No sea ice was present in the northern section between WP2 and WP3 (Fig. R3a, right y-axis) as shown by a surface temperature below the aircraft of about 3°C. In the southern section between WP4, WP5, WP6, and WP7, the surface contained both open water, scattered sea ice floes, and closed sea ice. This is demonstrated by surface temperatures ranging from about 3°C over open water close to WP4, over −1.8°C close to WP5 and WP6, to −15°C in the fjord close to WP7 (Fig. R3a, right y-axis). As mentioned above, easterly winds prevailed, resulting in off-ice flow, i.e., the areas probed inside the ABL over open water between WP4 and WP5 were located downwind of the sea ice (see Fig. R2b, Fig. R3a).

In both the northern and the southern section, particle number concentrations in the size range from 10 nm to 600 nm were increased inside the ABL compared to the free troposphere (Fig. R3b, c). In the northern section, no newly formed particles during the legs inside the ABL over completely ice-free ocean were present (Fig. R3b, c), as indicated by the absence of a prevailing distinct nucleation mode and rather low number concentrations in the size range from 10 nm to 20 nm ($N_{10-20} \approx 25\,\mathrm{cm}^{-3}$, ≈ 10 % of the total particle number concentrations measured in the range from 10 nm to 600 nm). One single PNSD measured at 9:30 UTC suggested the presence of a small nucleation mode with a maximum below 10 nm (Fig. R3b). However, because during this measurement a few scattered low-level clouds next to the aircraft were visible in the video data, but their distance to the aircraft could not be quantified and the leg was not carried out directly above/below the cloud top/base, this measurement is assigned to the unclassified category (see Fig. 2 and Fig. 3 in the main text and Sect. S3).

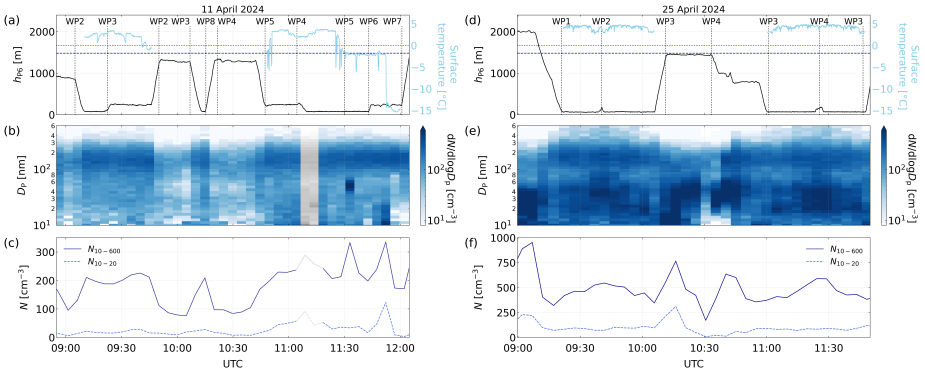

Figure R3: **(a)** P6 altitude during RF03 on 11 April 2024 (black line, left y-axis) as well as surface temperature (blue line, right y-axis). The dashed/dotted blue horizontal lines illustrate a surface temperature of 0°C and the freezing temperature of (salty) Arctic ocean water of −1.8°C. **(b)** MPSS-measured PNSD. The peak in the number concentrations N_{10-600} around 11:32 UTC results from a pollution plume (likely ship exhaust) generating particles with a mode around 60 nm. The measurements from 11:05 to 11:15 UTC are grayed out because the exhaust of the Polar 6 research aircraft was sampled during the turn at WP4. **(c)** Integral particle number concentrations in the total measured size range (10 nm $\leq D_p \leq$ 600 nm, solid line) and in the nucleation mode size range (10 nm $\leq D_p \leq$ 20 nm, dashed line), respectively. **(d)** P6 altitude during RF09 on 25 April 2024 (black line, left y-axis) as well as surface temperature (blue line, right y-axis). The dashed/dotted blue horizontal lines illustrate a surface temperature of 0°C and the freezing temperature of Arctic ocean water of −1.8°C. **(e)** MPSS-measured PNSD. **(f)** Integral particle number concentrations in the total measured size range (10 nm $\leq D_p \leq$ 600 nm, solid line) and in the nucleation mode size range (10 nm $\leq D_p \leq$ 20 nm, dashed line), respectively. Note the different y-axis between the subplots (c) and (f).

The number concentrations of particles in the nucleation mode size range from 10 nm to 20 nm increased by a factor of roughly two in the southern section inside the ABL over ocean with sea ice floes compared to the northern section (Fig. R3c). Here, $N_{10-20} \approx 50\,\mathrm{cm}^{-3}$, accounting for $\approx 20\,\%$ of the total particle number concentrations measured in the range from 10 nm to 600 nm. Several PNSDs showed a small but clear mode with a maximum below 10 nm (Fig. R3b, around 11:00, 11:15, 11:35, 11:45 UTC), indicative of the presence of newly formed particles. Interestingly, the largest nucleation mode (the highest peak in N_{10-20}) was observed directly at the transition between water with ice floes and closed sea ice in the fjord between WP6 and WP7 around 11:50 UTC (Fig. R3b). The median PNSDs for the legs inside the ABL are shown in Fig. R4 and will be discussed below.

On 25 April, four legs at altitudes of ≈ 67 m were carried out inside the ABL over open ocean (9:20 to 10:05 UTC and 11:00 to 11:45 UTC, Fig. R3d, left y-axis), i.e., with no sea ice present at all, as shown by a sea surface temperature of about 4°C (Fig. R3d, right y-axis). Additionally, one leg was flown at ≈ 1400 m in the free troposphere between WP3 and WP4 (10:10 to 10:35 UTC). Furthermore, one leg was carried out directly above the decreasing cloud top between WP3 and WP4 at altitudes between about ≈ 1000 m to ≈ 750 m (10:35 to 10:55 UTC). During the legs inside the ABL over open water, PNSDs showed no indications regarding the presence of a nucleation mode with a maximum below 10 nm, i.e., no indications for newly formed particles (Fig. R3e, f). In the free troposphere, around 9:00 UTC and 10:15 UTC, increased numbers of particles in the size range from 10 nm to 20 nm were present, and measured PNSDs indicated the presence of a pronounced nucleation mode. These newly formed particles in the free troposphere were associated with air masses from lower latitudes (Fig. R2e). However, we here focus on the measurements inside the ABL. The occurrence of newly formed particles in the free troposphere is discussed in detail in Sect. 3.2 in the main text.

A more detailed comparison of PNSDs measured during the legs inside the ABL over the Arctic ocean on 11 and 25 April is presented in Fig. R4. The accumulation mode concentrations are similar for all six median distributions depicted therein. Additionally, a sea salt mode was observed during the legs inside the ABL on 25 April (blue distribution in Fig. R4) due to sea spray aerosol (SSA) generation caused by strong winds (horizontal wind speeds of about $15\,\mathrm{m\,s}^{-1}$, strong fluctuations in vertical wind speeds; for a detailed case study on SSA fluxes on 25 April see Simon et al. 2025). The median PNSDs measured inside the ABL over completely ice-free ocean on 11 and 25 April featured no nucleation mode, i.e., no indications for the presence of newly formed particles were observed (blue and black distributions in Fig. R4). In contrast, PNSDs measured inside the ABL over water with sea ice floes indicated the presence of newly formed particles (red distributions in Fig. R4).

In summary, no clear indications for the occurrence of newly formed particles during measurements inside the ABL over open, completely ice-free ocean were found. However, a small number of newly formed particles were detected during measurements inside the ABL over ocean with sea ice floes, i.e., in the direct

vicinity and downwind of sea ice.

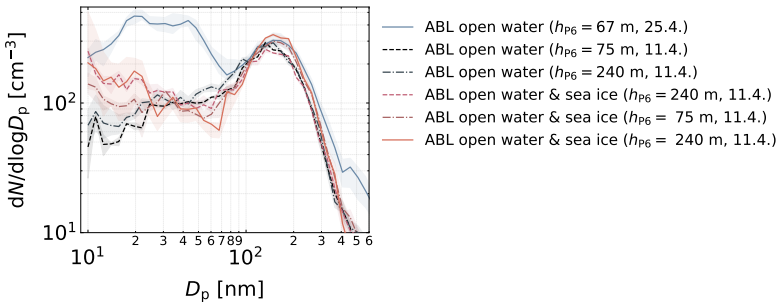

Figure R4: Median particle number size distributions during legs at constant altitude inside the ABL over completely ice-free open water (on 11 and 25 April, black and blue curves) and over ocean with both sea ice floes and open water areas present (on 11 April, red curves). The latter distributions over both open water and sea ice on 11 April are considered as indicative for newly formed particles. The shaded areas represent the interquartile ranges.

Because we now additionally discuss the observations of newly formed particles inside the ABL over ocean with sea ice floes, we have updated Fig. 3 in the main text (Fig. 2 in the previous version) as mentioned and shown above. Here, the median PNSDs which were originally depicted in a separate plot in the SI (Fig. S3 in the previous version) are now included in the panel Fig. 3a and removed from the SI. Additionally, we have updated the plot depicting the relative humidity and temperature conditions during the observations of newly formed particles to include the fourth category as well (Fig. S3 (Fig. S4 in the previous version)). For the latter, we used a refined calibration of the turbulence data and calculated the histograms based on the 100 Hz data instead of 1 Hz data. Please find the revised figure below (Fig. R5).

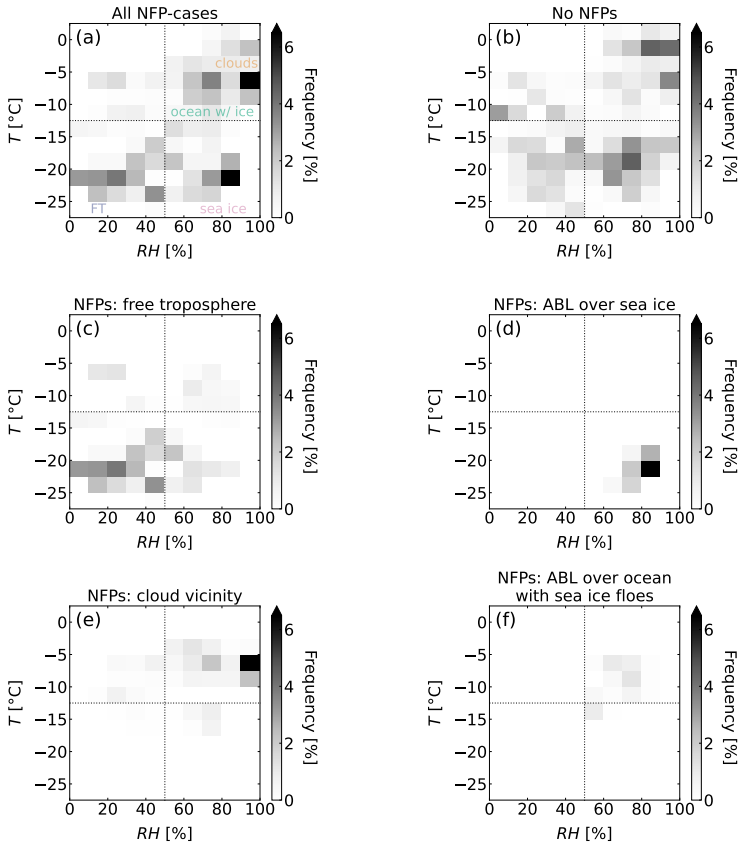

Figure R5: Temperature and relative humidity conditions during **(a)** newly formed particle-cases (NFP-cases) and **(b)** non-NFP-cases. The labels "FT", "sea ice", "clouds", and "ocean w/ ice" in panel (a) denote that within the corresponding quadrant, the measured temperature and relative humidity values are predominantly associated with newly formed particles observed in the free troposphere, the ABL over sea ice, cloud vicinity, and the ABL over ocean with sea ice floes, respectively. The second and third row depict temperature and relative humidity during the occurrence of newly formed particles **(c)** in the free troposphere, **(d)** the ABL over sea ice, **(e)**, the vicinity of clouds, **(f)** and the ABL over ocean with sea ice floes.

iii. The manuscript would benefit from a bit more discussion on sources of precursors. The seasonal aspect should be considered, the observations were made in early spring, has biological productivity in the ocean (and presumed production of precursor gases) begun? Have you checked e.g satellite products? The main conclusions should also be set in this context for future reference.

Thank you for this nice suggestion. Precursors and their sources were already discussed in the case study on newly formed particles in the ABL over sea ice (Sect. 3.3), as this research flight comprises measurements (at low altitudes) during our campaign for which emissions of precursors from the surface offer a plausible explanation for the occurrence of newly formed particles in the target region.
Additionally, motivated by this remark as well as by the reviewer's comment #15, we used Copernicus Global Ocean Colour data (Gohin et al. 2002; Hu et al. 2012) to investigate potential precursor emissions from the ocean surface during the measurements inside the ABL over ocean. Briefly, we have found no indications that significant biological productivity in the open ocean occurred in the target regions for measurements over completely ice-free ocean (on April 11 and 25). However, during the measurements inside the ABL on 11 April conducted over ocean with ice floes (with the surface containing both sea ice and open water), the data suggest that a phytoplankton bloom occurred in the target region. We are grateful for the suggestion and have included a detailed discussion on this analysis in the SI in Sect. S5.3 (i.e., right after the discussion of the PNSD measurements over ocean on 11 and 25 April). Please find this section below (lines 168 to 210):

S5.3 Chlorophyll-a concentration

We utilized satellite-derived Copernicus Global Ocean Colour data (Gohin et al. 2002; Hu et al. 2012) to carry out an analysis with respect to the chlorophyll-a (chl-a) mass concentration in the sea water during the measurements inside the ABL over ocean on 11 and 25 April. Here, chl-a is treated as a proxy for phytoplankton biomass (Siegel et al. 2013). It is well known that the aerosol precursor dimethylsulfide (DMS) results from the breakdown of dimethylsulfoniopropionate (DMSP), which is produced by phytoplankton (e.g., Simó 2001; Willis et al. 2017). Thus, we assume that the chl-a concentrations are suited to investigate the potential for the release of DMS from the ocean surface to the atmosphere. In the Arctic at Ny-Ålesund, for example, Beck et al. (2021) showed a good agreement between measured chl-a and DMSP concentrations in seawater, which coincided with a rise in sulfuric acid (SA) and methane sulfonic acid (MSA), both of which are oxidation products of DMS, and the occurrence of new particle formation (NPF) and growth events.
Compared to the background values close to zero, elevated chl-a concentrations are visible in the southern section of the target region on 11 April (Fig. R6a) where both open water and sea ice were present (Fig. R2a, R3a), suggesting that in the sea water in the area probed a phytoplankton bloom occurred. Furthermore, the data show no or only slightly increased chl-a concentrations in the northern section (Fig. R6a).

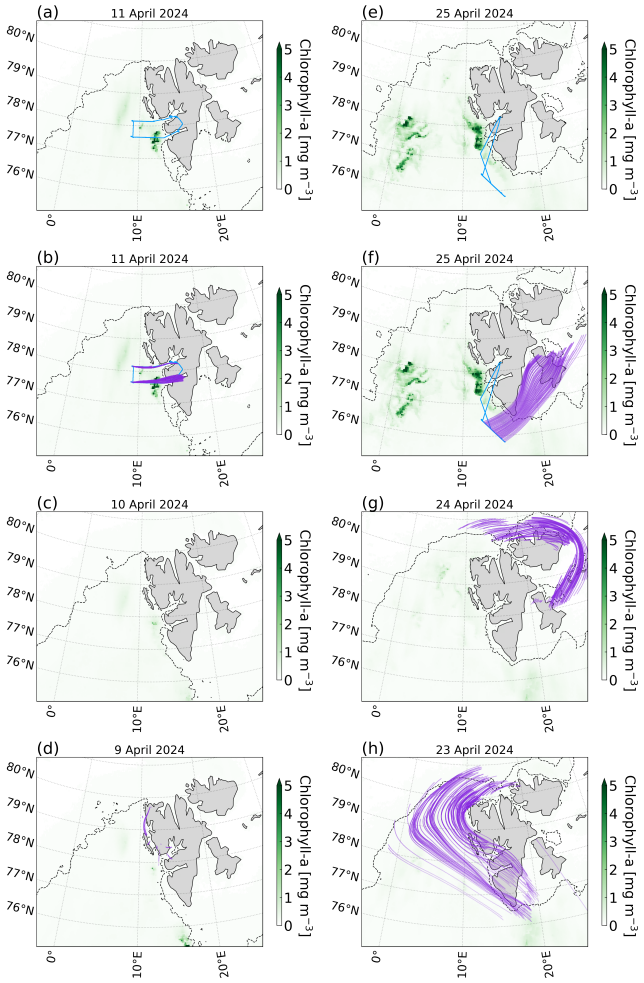

Figure R6: Flight track (blue), backward trajectories inside the ABL (violet), and chlorophyll-a mass concentrations (green) based on Copernicus-GlobColour data (Gohin et al. 2002; Hu et al. 2012) for the research flights featuring measurements inside the ABL over ocean on 11 and 25 April. The left column depicts the chlorophyll-a concentrations for 11 April 2024 and the two preceding days. Similarly, the right column depicts the chlorophyll-a concentrations for 25 April 2024 and the two preceding days. For convenience, only the respective flight tracks and chlorophyll-a concentrations for the days of the research flights are shown in the first row in panels (a) and (e). In the panels (b), (d), (d), (f), (g), and (h), the backward trajectories during the corresponding days are depicted as well. Only trajectories inside the ABL are shown, i.e., if no trajectory data are shown such as in panel (c) or gaps in the trajectories are visible such as in (d), (g), and (h), the trajectories did not reside inside the ABL at these geographical positions. The dashed black line in all subplots illustrates the sea ice extent based on 15 % sea ice concentration (Spreen et al. 2008).

Fig. R6b depicts the backward trajectories inside the ABL on 11 April. We restrict the following discussion to air masses which resided inside the ABL since we expect that only those air masses could have picked up precursor gases from the ocean surface, whereas an influence of emissions from the surface on free tropospheric air masses is unlikely. Consequently, only the parts of the trajectories inside the ABL are shown in Fig. R6b-d, f-h. As mentioned above, easterly winds prevailed in both the northern and the southern section (Fig. R6b). The air masses sampled in the northern section, for which no newly formed particles were observed, did not pass over ocean surfaces featuring enhanced chl-a concentrations (Fig. R6b). In contrast, in the southern section, measurements in the direct vicinity of ocean waters with increased chl-a were conducted, as well as measurements downwind of these areas indicative of a phytoplankton bloom. On the two previous days, the air massed resided completely (10 April, Fig. R6c) and overwhelmingly outside of the ABL (9 April, Fig. R6d), limiting the possibility for precursor uptake from the (ocean) surface on those days. This suggests that the presumed emission of precursors, potentially leading to NPF and the occurrence of newly formed particles in the southern section of the target area on 11 April, occurred rather locally.

In the area probed on 25 April, the chl-a data showed no indications for a phytoplankton bloom (Fig. R6e). Similarly, in the areas where backward trajectories resided inside the ABL during 25 April and the two preceding days, no elevated chl-a concentrations were present in the sea water (Fig. R6f-h).

In summary, no evidence for a phytoplankton bloom in the northern section of the area probed on 11 April and in the area probed on 25 April was found based on the satellite data. However, in the southern section of the target region investigated on 11 April, the chl-a data suggest that a phytoplankton plume occurred. This might explain why newly formed particles were found inside the ABL over ocean with sea ice floes above and downwind of the plume area and the sea ice, but no newly formed particles were detected during the measurements inside the ABL over completely ice-free ocean. However, the occurrence of newly formed particles between WP6 and WP7 (Fig. R3a-c) upwind of the plume area on April 11 was likely not affected by the chl-a plume as the wind direction would prevent the eastward transport of possibly emitted precursors (Fig. R6b). Due to the absence of gas phase measurements, we cannot ultimately resolve if the potential release of precursors such as DMS from the sea surface and its possible participation in NPF might have contributed to the occurrence of newly formed particles on 11 April. Hence, we can only speculate that the presence of newly formed particles over ocean with sea ice floes on this day might be related to emissions of precursors from the open water or the sea ice, or both. Another potential explanation has been initially proposed by Leck and Bigg (1999, 2010) who suggested that the fragmentation of marine organic polymer gels led to the appearance of particles in the nucleation mode size range. Such events often involved the dissipation of cloud or fog droplets, emitting particles with sizes of a few nanometers to the atmosphere (Leck and Bigg 1999, 2010; Karl et al. 2013; Leck et al. 2025).

iv. Figure 2d in its current form is highly misleading. Since the measurement time is not evenly distributed across altitudes, nor concurrent in time or space, these profiles do not make sense. If you want to show vertical distributions of number concentrations, I think it should be done in a way where the reader can distinguish between flights and where the number of observations (or flighttime) is reflected. However, the figure is barely mentioned and not discussed in the text, so it could also be omitted.

Thank you for this critical comment. We agree that showing such a profile might indeed lead to misinterpretation since the measurement times were not evenly distributed over the investigated altitudes, geographical locations, and synoptic conditions, as also discussed in the original manuscript. Consequently, as suggested by the reviewer, we have removed Fig. 2d and corresponding references in the main text from the manuscript. The revised figure can be found above in our response to the reviewer's comment #ii (Fig R1).

Detailed comments

1. Line 34: I think a correlation should rather be referred to as indirect evidence.

Thanks for noting this! We agree with the reviewer and have changed the sentences to (lines 34 to 35):
There is indirect evidence reported by Dall'Osto et al. (2017, 2018) who found a correlation between an increase of NPF events and decreasing sea ice extent.

2. Line 36: See also Heslin-Rees et al, 2020 (https://doi.org/10.5194/acp-20-13671-2020) who showed that changes in transport patterns into the Arctic increased the marine influence on the aerosol population, rather than sea ice retreat.

Thank you very much for pointing us to this interesting publication! We have included a corresponding reference and sentence (lines 38 to 39):
Similarly, Heslin-Rees et al. (2020) showed that changed air circulation patterns resulted in an increased marine influence on the aerosol population at the Arctic Zeppelin station (Svalbard).

3. Line 59: I interpret Pilz et al (2024) in the exact opposite way; free tropospheric sources of CCN were more common. Please revisit.

Yes, indeed, Pilz et al. (2024) reported enhanced free tropospheric particle sources, as we also discussed more extensively in lines 200 - 206 of the original manuscript (lines 284 to 290 in the revised manuscript). Thank you for noting this! In response to this comment, but also due to the reviewer's comment #10,

we have revised this section of the introduction, which now reads (lines 49 to 57):

Consequently, to observe and analyze NPF, airborne measurements with balloons, helicopters, or aircraft appear indispensable (Garrett et al. 2002; Weber et al. 2003; Engvall et al. 2008; Khosrawi et al. 2010; Kupiszewski et al. 2013; Willis et al. 2016; Burkart et al. 2017; Willis et al. 2017; Pilz et al. 2024; Pohorsky et al. 2026). Despite these growing efforts to conduct vertically-resolved measurements on aerosol particles in general, and NPF in particular, data on the vertical distribution of NPF in the Arctic are still scarce (e.g., Burkart et al. 2017; Schmale et al. 2021; Pohorsky et al. 2026). Hence, the relative importance of the ABL and the free troposphere as sources of newly formed particles remains elusive, with a number of studies reporting on the predominant occurrence of NPF close to the surface (Kupiszewski et al. 2013; Willis et al. 2016; Burkart et al. 2017; Willis et al. 2017, 2018), while other measurements detected NPF in the free troposphere as well (Weber et al. 2003; Khosrawi et al. 2010).

4. Lines 82-83: Please add whether the aerosol data have also been corrected for diffusion and inertial deposition or not.

Thank you for raising this point! We had calculated particles losses due to diffusion and inertial deposition using the Particle Loss Calculator (Von der Weiden et al. 2009), but erroneously not applied the correction factors to the data shown in the original manuscript. We have now corrected all PNSD data for particle losses in the sampling tubing from the aircraft's aerosol inlet to the MPSS. The procedure is outlined in a separate section in the SI of the revised manuscript (Sect. S2). The transmission efficiencies as a function of the particle diameter can be found below in Fig. R7.

The only changes made in the main text of the manuscript refer to one additional sentence in the methods section describing that we corrected for losses, and updated values of particle number concentrations in the size ranges from 10 nm to 20 nm (N_{10-20}) and 10 nm to 600 nm (N_{10-20}), respectively, as well as of the fraction N_{10-20}/N_{10-600} reported for each case study. Overall, neither our analysis nor our conclusions are affected in any substantial way but only minor quantitative modifications have been made. We have now also updated all figures displaying PNSD data with the loss-corrected values. Please find the revised sections below.

Methods (Sect. 2.2 in the main text, lines 89 to 92):

Particle losses inside the sampling tubing from the aerosol inlet to the MPSS due to diffusion, sedimentation, turbulent inertial deposition, and inertial deposition in bends and contractions have been corrected for based on calculations utilizing the particle loss calculator (Von der Weiden et al. 2009, Sect. S2).

SI (lines 5 to 31):

S2 Correction for particle losses

To characterize particle losses in the sampling line from the aerosol inlet of

the Polar 6 aircraft to the mobility particle size spectrometer (MPSS), we calculated the transmission efficiencies ($\eta = \eta(D_\mathrm{p})$, where D_p is the particle diameter) in the tubing utilizing the Particle Loss Calculator (Von der Weiden et al. 2009). The particle loss calculator includes losses due to diffusion, sedimentation, turbulent inertial deposition, and inertial deposition in bends and contractions. To approximate the aerosol particles' parameters, we assumed a density of $1.77\,\mathrm{g\,cm^{-3}}$, representing the density of ammonium sulfate particles, and a shape factor of 1, corresponding to spherical particles (Seinfeld and Pandis 2006). As discussed in Sect. 2.2 of the main text, the flow in the sampling tubing is manually adjusted depending on the true airspeed of the aircraft (typically $tas \approx 60\,\mathrm{m\,s^{-1}}$ during flight section at low altitudes in the target regions, and $tas \approx 90\,\mathrm{m\,s^{-1}}$ during flight sections at high altitudes, e.g., transit flights). This procedure enables near-isokinetic sampling (Jurányi et al. 2025). Based on the corresponding flow recordings as well as pressure and temperature measurements of the MPSS throughout the campaign, for the loss calculations, pressure and temperature in the sampling line are set to the respective median values for measurements at high and low altitudes: $p_\mathrm{med} = 761\,\mathrm{hPa}$, $T_\mathrm{med} = 290\,\mathrm{K}$ for $tas \approx 90\,\mathrm{m\,s^{-1}}$, and $p_\mathrm{med} = 987\,\mathrm{hPa}$, $T_\mathrm{med} = 298\,\mathrm{K}$ for $tas \approx 60\,\mathrm{m\,s^{-1}}$. The calculations yield the transmission efficiency curves shown as solid lines in Fig. R7. To estimate the uncertainty in the transmission efficiencies associated with the median settings, we show the transmission efficiencies calculated for the 10th and 90th percentiles of the pressure and temperature distributions as shaded areas in Fig. R7 as well. Here, the respective parameters are set to $p_{10} = 678\,\mathrm{hPa}$, $T_{10} = 286\,\mathrm{K}$, $p_{90} = 905\,\mathrm{hPa}$, $T_{90} = 296\,\mathrm{K}$ for $tas \approx 90\,\mathrm{m\,s^{-1}}$, and $p_{10} = 862\,\mathrm{hPa}$, $T_{10} = 293\,\mathrm{K}$, $p_{90} = 1030\,\mathrm{hPa}$, $T_\mathrm{med} = 300\,\mathrm{K}$ for $tas \approx 60\,\mathrm{m\,s^{-1}}$.

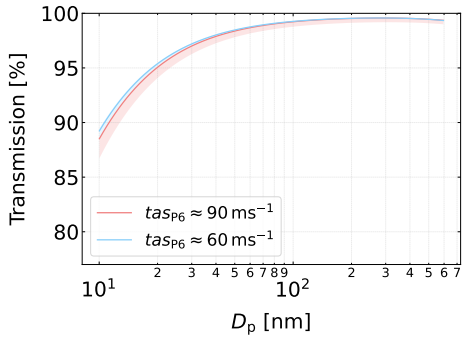

Figure R7: Particle size-dependent transmission efficiencies in the sampling tubing from the aerosol inlet to the MPSS for the two typical true airspeeds of the Polar 6 aircraft during the campaign.

For both typical true airspeeds of the aircraft, the transmission efficiencies calculated for the median pressure and temperature values range from $\approx 89\,\%$ at 10 nm over $\approx 95\,\%$ at 20 nm to $> 99\,\%$ at 100 nm (solid lines in Fig. R7). The values decrease towards smaller particle diameters to their minimum values at

10 nm as diffusional losses increase with decreasing particle size (Baron and Willeke 2001, Fig. R7). Accordingly, we correct the MPSS-measured particle number size distribution (PNSD) data for particles losses in the sampling tubing by size-dependent correction factors ($c(D_\mathrm{p}) = 1/\eta(D_\mathrm{p})$) ranging from close to 1 to 1.12. Considering the transmission efficiencies calculated for the 10th and 90th percentiles, maximum differences are $\approx 1\,\%$ for $tas \approx 60\ \mathrm{m\,s^{-1}}$ and $\approx 2\,\%$ for $tas \approx 90\ \mathrm{m\,s^{-1}}$ compared to the corrections based on the medians of the pressure and temperature distributions. We consider these errors to be negligible and apply the correction factors based on the median values to all PNSD data.

Sect. 3.2 (Newly formed particles in the free troposphere, lines 214 to 220):
Detected particle number concentrations in the size range from 10 nm to 20 nm reached on average values of about $N_{10-20} \approx 778\ \mathrm{cm^{-3}}$ [previously 714 $\mathrm{cm^{-3}}$], which constituted about 68 % [previously 66 %] of the total particle number concentration measured in the range from 10 nm to 600 nm [...]. For the time period 7:40 to 8:19 UTC, encountered PNSDs featured no distinct nucleation mode, and number concentrations in the size range from 10 nm to 20 nm were substantially lower ($N_{10-20} \approx 27\ \mathrm{cm^{-3}}$, 17 % [previously 25 $\mathrm{cm^{-3}}$, 16 %] of the total MPSS-measured number concentration [...].

Sect. 3.3 (Newly formed particles inside the atmospheric boundary layer over sea ice, lines 319 to 322):
Mean number concentrations in the nucleation mode size range reached $N_{10-20} \approx 206\ \mathrm{cm^{-3}}$ [previously 191 $\mathrm{cm^{-3}}$] (time period 10:03 to 10:39 UTC), $N_{10-20} \approx 185\ \mathrm{cm^{-3}}$ [previously 170 $\mathrm{cm^{-3}}$] (11:08 to 11:22 UTC), and $N_{10-20} \approx 125\ \mathrm{cm^{-3}}$ [previously 114 $\mathrm{cm^{-3}}$](12:17 to 12:40 UTC), accounting for about 40 %, 38 %, and 31 % [previously 39 %, 37 %, and 30 %] of the total MPSS-measured aerosol particle number concentrations.

Sect. 3.4 (Newly formed particles in the vicinity of clouds, lines 399 to 402):
The presence/absence of particles in the size range from 10 nm to 20 nm is also clearly visible in Fig. 11c, with particles in the size range below 20 nm contributing approximately 67 %, 60 %, and 9 % [previously 65 %, 59 %, and 9 %] to the total aerosol particle number concentrations at cloud top, cloud base, and in the free troposphere, respectively.

5. Line 114: I suggest removing the word frequently. In line 121 you state it was 24 % of the flight time that you identified NFP, whether that is frequent or not is not obvious but subjective.

We have changed the title of this subsection to (line 129):
Geographical and vertical distribution of newly formed particles

6. Line 129: "Newly formed particles detected in the free troposphere account for the majority of our observations" is a misleading statement and can be

used to misquote this study. 45 % of all PNSD featuring newly formed particles is actually not even the majority of those cases (that would require > 50 %), and definitely not the majority of your observations.

Agreed. We have changed the sentence to (lines 155 to 156):
About 45 % of all PNSDs featuring newly formed particles were detected in the free troposphere, corresponding to approximately 4 hours and 40 minutes of measurements (Fig. 3 a, b) and distances flown of ≈ 1460 km.
Please find the full revised paragraph in our response to the reviewer's comment #7 below.

7. Line 130-132: The percentages presented here lack context, they do not constitute a result without knowing how much flight time was spent in each category.

As outlined in the response to the reviewer's comment #ii, the measurement times per category can only be (roughly) quantified, based on reanalysis data. These measurement times are now provided in the SI in Sect. S3. Please find this SI section in our response to the reviewer's comment #ii. Specifically with respect to the paragraph the reviewer refers to here, the revised version now reads (lines 156 to 165):
About 45 % of all PNSDs featuring newly formed particles were detected in the free troposphere, corresponding to approximately 4 hours and 40 minutes of measurements (Fig. 3a, b) and distances flown of ≈ 1460 km. About 16 % (≈ 1 hour and 40 minutes, ≈ 350 km) of all cases featuring newly formed particles were found in the ABL over sea ice, as well as 6 % (≈ 40 minutes, ≈ 130 km) in the ABL over ocean with sea ice floes. Approximately 25 % (≈ 2 hours and 40 minutes, ≈ 630 km) of all cases were observed in the vicinity of clouds(Fig. 3a, b). For approximately 8 % (≈ 50 minutes, ≈ 200 km) of all PNSDs featuring newly formed particles, the prevailing conditions could not unambiguously be identified, i.e., these cases could not clearly be assigned to one of the categories or to another separate category (Fig. 3b, c). They include flight sections during which clouds were present, but in contrast to the newly formed particles classified as in the vicinity of clouds, we did not carry out dedicated legs directly at cloud top or base, respectively. Also included are flight legs for which it could not unequivocally be determined if the legs were conducted inside or outside the ABL (see Sect. S3).

We are grateful for the comment and have additionally updated Fig. 3 and now only include the duration and frequency of NFP-occurrence per altitude, as well as the total measurement time per altitude. The percentages the reviewer refers to here, i.e., the fraction of all NPF-observations for each category are not shown anymore. Please find the revised figure below (Fig. R8) and also see our response to the reviewer's comments #8 and # ii. Note that Fig. R8 (corresponding to Fig. 3 in the main manuscript) is the same as Fig. R1 and shown here again, for convenience.

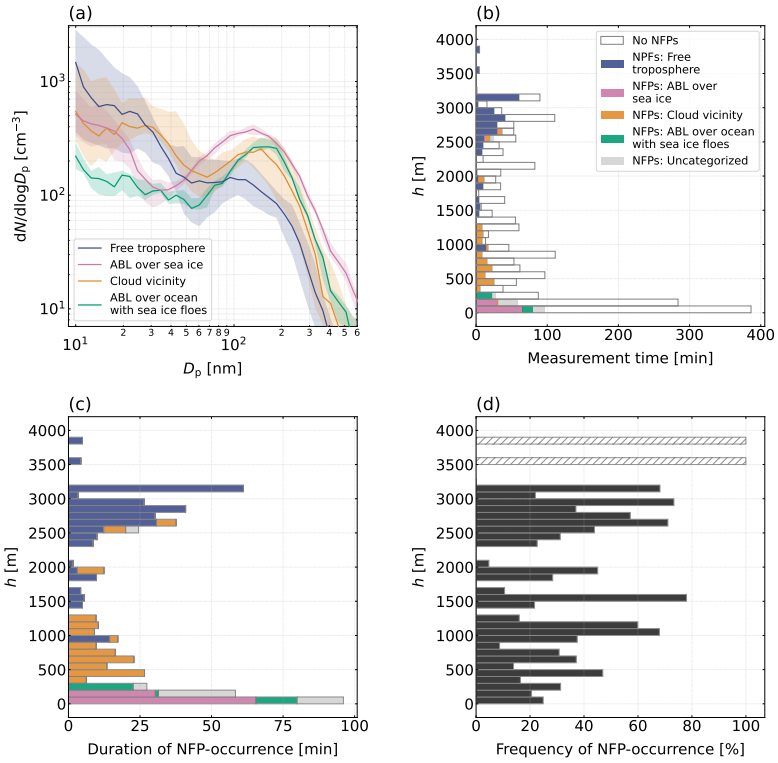

Figure R8: **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and inside the ABL over ocean with ice floes (Sect. S5). **(b)** Total measurement times per altitude (grouped into height bins with a width of 100 m) showing the absence of newly formed particles as well as times during which newly formed particles were detected for the different environments. **(c)** For convenience, this subplots shows the duration of newly formed particle-occurrence (NFP-occurrence) per measurement altitude again for the different categories, i.e., the same NFP data as in the first panel. The NFP data here and in panel (a) are shown as stacked bars. Note the different range of the x-axis compared to (a). **(d)** Relative frequency of NFP-occurrence per measurement altitude (irrespective of category), i.e., NFP-occurrence divided by total measurement time. The hatched bars at altitudes higher than 3500 m indicate that here only one full PNSD measurement was conducted within each altitude bin, while below 3200 m on average 14 MPSS scans were carried out at each altitude, with a maximum of 79 scans between 60 and 100 m (Fig. R8a).

8. Figure 2c: How many scans do the non-hatched bars represent? Preferably add the number for each bar, or at least include a minimum and mean value.

We thank the reviewer for this suggestion! We found that including the number of scans at each bar overcrowded the figure quite considerably, while at the same time not providing a lot of additional meaningful insights. However, of course the measurement time at each altitude is very important. We have therefore both modified Fig. 3 (Fig. 2 in the previous manuscript) to feature the total measurement time, and included the mean number of scans and a maximum value in the description. The updated figure now shows the total measurement time per altitude in panel b, together with the times during which NFPs were observed, which are color-coded by the respective category. Fig. 3c, d show the duration and relative frequency of newly formed particle-occurrence as function of altitude as in the previous manuscript in Fig. 3b, c. Please find the revised figure above in our response to the reviewer's comment #7 (Fig. R8).

9. Line 142-146: I believe this is the only reference to figure 2d. Mentioning the peaks at 1000 and 3000 m should be followed by discussion, how much observational time does this reflect? Are these peaks representative? In figure 2b it looks like the NPF-occurrence was not very long at these heights.

We thank the reviewer for raising this point. Indeed, the profile shown here could potentially lead to misinterpretation since the measurement times at different altitudes were neither distributed uniformly over the target regions and synoptic conditions nor vertically homogeneous. We have removed Fig. 2d as well as lines 142 - 146 from the manuscript. On this note, please see also our response to the reviewer's comment #iv. The revised figure Fig. 3 (Fig. 2 in the originally submitted manuscript) is depicted above in our response to the reviewer's comment #7 (Fig. R8).

10. Line 195: Were researchers led to believe that? In recent years, a lot of effort has been put into making vertical measurements of aitken and nucleation mode particles (e.g Pohorsky et al (preprint at https://egusphere.copernicus.org/preprints/2026/egusphere-2026-1068/), need for vertical measurements discussed e.g in Schmale et al, 2021). Would this be the case if the research community believed it wasn't important? Near-surface measurements are more accessible, and have therefore been conducted to a higher degree.

Thank you for this helpful comment. Here, we specifically referred to the following statements made in the publications by Willis et al. (2018) and Dall'Osto et al. (2017, 2020), which we also cited in lines 195-196 in the original manuscript:

- "Previous observations showed new particle formation events taking place

at high altitudes including the free troposphere[30] [Wiedensohler et al., 1996] as well as in the boundary layer near the surface[31][Ström et al., 2009]. Recent vertical profiles have revealed the latter as a more plausible and common source[32,33] [Kupiszewski et al., 2013; Burkart et al., 2017]".
Dall'Osto et al. (2017)

- "Aircraft measurements of ultrafine particles indicate that their formation predominantly occurs in the Arctic lower troposphere (Burkart, Willis, et al., 2017; Willis et al., 2016), though the free troposphere also contributes (Igel et al., 2017; Weber et al., 2003)."
Willis et al. (2018)

- "A number of observations have shown that new particle events can take place at high altitude, including in the free troposphere (Wiedensohler et al., 1996). However, recent vertical profiles taken in the last decade have revealed that nucleation events in the boundary layer near sea ice and open water regions may be a more plausible and much common dominating source."
Dall'Osto et al. (2020)

However, the reviewer is right that recently several studies have been conducted to investigate the vertical distribution of nucleation and aitken mode particles, and that surely not all researchers were led to believe that NPF occurs mainly close to the surface. The reviewer rightfully raises the concern that such a strong wording could lead to misinterpretation and incorrectly imply a consensus in the community that free tropospheric NPF was of minor importance. We are grateful to the reviewer's comment which has helped us to improve the manuscript. Accordingly, we have rephrased the discussion on free tropospheric newly formed particles in Sect. 3.2, the abstract, as well as the part of the introduction in which we discuss previous airborne measurements (see also our response to the reviewer's comment #3). Please find the revised paragraphs below.

Abstract (lines 1 to 11):
New particle formation (NPF) can impact the Arctic radiative energy budget since this region is particularly sensitive to changes in aerosol particle and cloud condensation nuclei concentrations. Prior measurements have predominantly investigated NPF in the Arctic atmospheric boundary layer (ABL), while vertically-resolved studies on newly formed aerosol particles remain scarce. Here, we present an analysis which suggests that NPF may take place throughout the entire lower Arctic troposphere. We have reached this conclusion by analyzing aerosol particle number size distribution data collected during a springtime aircraft campaign (BACSAM II) in the vicinity of Svalbard, the Fram Strait, and northern Greenland. We detected newly formed aerosol particles over distances of hundreds of kilometers, at various altitudes ranging from about 60 m to 3900 m, and identified four atmospheric scenarios for their occurrence: newly formed particles in the free troposphere, in the ABL over sea ice, in the

vicinity of clouds, and in the ABL over ocean with sea ice floes. Our results indicate that regional Arctic atmospheric processes as well as long-range transport may play key roles in the formation of new particles. In particular, we deduce that one possible mechanism leading to free tropospheric NPF could be associated with warm-air intrusions.

Introduction (lines 49 to 57):
[...] Consequently, to observe and analyze NPF, airborne measurements with balloons, helicopters, or aircraft appear indispensable (Garrett et al. 2002; Weber et al. 2003; Engvall et al. 2008; Khosrawi et al. 2010; Kupiszewski et al. 2013; Willis et al. 2016; Burkart et al. 2017; Willis et al. 2017; Pilz et al. 2024; Pohorsky et al. 2026). Despite these growing efforts to conduct vertically-resolved measurements on aerosol particles in general, and NPF in particular, data on the vertical distribution of NPF in the Arctic are still scarce (e.g., Burkart et al. 2017; Schmale et al. 2021; Pohorsky et al. 2026). Hence, the relative importance of the ABL and the free troposphere as sources of newly formed particles remains elusive, with a number of studies reporting on the predominant occurrence of NPF close to the surface (Kupiszewski et al. 2013; Willis et al. 2016; Burkart et al. 2017; Willis et al. 2017, 2018), while other measurements detected NPF in the free troposphere as well (Weber et al. 2003; Khosrawi et al. 2010). [...]

Sect. 3.2 (Newly formed particles in the free troposphere, lines 276 to 303):
The presence of newly formed particles in the Arctic free troposphere has occasionally been reported before (Weber et al. 2003; Khosrawi et al. 2010). For example, Weber et al. 2003 conducted systematic aircraft measurements in the free troposphere of the Canadian Arctic from January to May 2000. During one of 38 research flights, they observed one clear NPF event at an altitude of approximately 4200 m over the Canadian Arctic Archipelago. During the other 37 flights of their campaign, no indications for NPF occurring at high altitudes were found. Similarly, Khosrawi et al. 2010 detected one NPF event in the upper free troposphere at an altitude of 7000 m over Storfjoden (Svalbard). Below 4000 m, nucleation mode particles were reported to occur quite frequently during their campaign centered around Svalbard in May and June 2004. Yet, no further altitude-resolved information or discussion discriminating between ABL and free tropospheric NPF was provided, except for one case study, where in addition to the NPF event at 7000 m, nucleation mode particles were found inside the ABL. Analyzing measurements conducted with a tethered balloon system, Pilz et al. 2024 reported enhanced number concentrations of aerosol particles in the size range from 12 nm to 150 nm concurrently with low concentrations of particles larger than 150 nm in the summertime Central Arctic during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) expedition. The authors hypothesized that NPF taking place aloft lead to these elevated concentrations of smaller particles but were unable to constrain potential sources and associated processes due to a limited maximum measurement altitude of 1000 m and because of missing PNSD measurements. Utilizing heli-

copter (Kupiszewski et al. 2013) and aircraft measurements (Willis et al. 2016; Burkart et al. 2017; Willis et al. 2017), a number of studies reported generally lower concentrations of nucleation mode particles in the Arctic free troposphere compared to the ABL. In these studies, however, several NPF events were observed near the surface.

Similar to the previous observational results, our data show that newly formed particles occurred in the free troposphere. Notably, our observations suggest that these particles prevailed over distances of several hundreds of kilometers. Whether the formation of clouds and thus cloud radiative forcing could be affected by the presence of newly formed particles in the free troposphere following their potential growth to CCN sizes, remains to be assessed and is both beyond the scope of this article and not feasible with the present dataset. In this regard, e.g. Leck and Bigg (1999) and Bigg and Leck (2001) described that during their summertime campaign over the central Arctic Ocean, highly stable conditions prevented mixing between the free troposphere and the ABL. Pointing toward the importance of free tropospheric particles for cloud formation, however, Pohorsky et al. (2026) reported that free tropospheric sources of CCN were required to explain measured cloud droplet number concentrations. This is in line with a large-eddy simulation study by Igel et al. (2017) which demonstrated that elevated aerosol particle layers in the free troposphere could be entrained into the cloud-topped ABL by cloud-induced mixing.

Thank you as well for pointing us to the recent publication by Pohorsky et al. (2026). We have included a reference to this paper in the introduction (see above), in the discussion on free tropospheric newly formed particles (see above), and in the section on newly formed particles in the vicinity of clouds:

Sect. 3.4 (Newly formed particles in the vicinity of clouds, lines 430 to 433):
In the Arctic, enhanced concentrations of particles in the size range from 20 nm to 300 nm at the top of stratus clouds were previously observed by Garrett et al. 2002, who suggested that recent particle formation at cloud top lead to these high concentrations. Similarly, a burst of nucleation mode particles within 50 m of the top of an Arctic low-level cloud was recently detected by Pohorsky et al. (2026).

11. Line 197-207: The information given here is highly relevant, but I question the conclusion of this section. The cited Weber study observed one NPF events during one in 38 flights, at 4200m. Why do you say that your results (in contrast?) indicate that NPF occurs frequently in the Arctic FT, based on the 11 flights of which two above 3200m if I understand correctly? If you had not encountered NFP during these flights, would you have concluded that NPF may not occur at that height at all? I don't think so. Similarly, you can also not claim that it happens all the time because you observed it. Overall, I think that a discussion on where NPF "primarily occurs" in the Arctic can not be settled based on this limited set of measurements.

Thank you for this comment. The reviewer is of course right that a discussion where NPF primarily occurs cannot be settled here, which was not our intention. We have rephrased and softened the discussion in this paragraph to avoid this impression. Please find the revised paragraph above in the response to the reviewer's comment #10 (lines 276 to 303 in Sect. 3.2).

12. Line 244: See also Khadir et al, 2023 (https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2023GL104325) for the link between NPF and precipitation found at Zeppelin Observatory. To me, it looks like something happens specifically around 24-30 hours back for the NFP trajectories, particularly in panel 5b,d,e,f,g and i. I think this could be investigated or at least discussed further.

Thank you a lot for pointing us to this very interesting publication! We agree that it likely played an important role that, during the time period of about 24 to 48 hours prior to arrival, clouds precipitated and dissolved, removing large aerosol particles and possibly releasing precursors, which could subsequently participate in NPF and lead to the occurrence of newly formed particles at the high altitudes observed during our campaign. We have extended the discussion in the respective paragraph. Please find the revised version below:

Lines 240 to 307 in Sect. 3.2
About 60 hours before arrival, temperatures of air masses linked to newly formed particles started to decrease, and between 24 to 48 hours prior to arrival, specific humidities decreased as well, while for the trajectories featuring no newly formed particles, the specific humidities remained rather constant throughout the last five days. Consequently, air masses associated with newly formed particles became colder and less humid compared to air masses showing no signs of free tropospheric newly formed particles. The latter, i.e., the decrease of specific humidity, might be caused by the formation of precipitating clouds, as already mentioned in the case study above. Considering the 75th percentile of the specific humidity in addition to the median values, the analysis suggests that some precipitation already set in up to roughly 100 hours prior to arrival for trajectories connected to newly formed particles. About 80 % of all backward trajectories linked to free tropospheric newly formed particles were in contact with clouds on their way to the measurement location during the previous five days, as well as about 70 % of the backward trajectories featuring no newly formed particles (Fig. 6i). However, no clouds were present during the last 24 hours before the measurement for the air masses featuring newly formed particles. For approximately 50 % of the respective trajectories the last contact with clouds occurred between about 24 to 48 hours prior to arrival(Fig. 6i). The decrease in specific humidity during the same time period (Fig. 6f) already mentioned above indicates that the removal of clouds, at latest 24 hours prior to arrival, could (partly) be caused by precipitation.
[...]
The above findings imply a potential role for clouds in the transport and pos-

sibly transformation of precursors, e.g., through releasing precursor gases from evaporating cloud droplets (Weber et al. 2001; Wehner et al. 2015). Precipitation caused by these clouds might have also increased the probability for NPF to occur by removing large aerosol particles, thus decreasing the pre-existing particle surface area and consequently reducing scavenging of gaseous precursors and newly formed particles (Perry and Hobbs 1994; Weber et al. 2001). Interestingly, already Khadir et al. (2023) observed a consistently positive correlation between nucleation mode particles and precipitation rate within the air mass during the transport to Zeppelin research station from about 24 to 96 hours prior to arrival, and mostly negative correlations between precipitation rate and nucleation mode particles in the last 12 hours. These results were found for data recorded from March to May, i.e., for a similar season as for our campaign, and covered the years from 2010 to 2019. Although with our dataset we cannot robustly identify significant correlations similar to Khadir et al. (2023), our analysis likewise suggests that precipitating clouds (occurring until about 24 hours prior to the measurement) might create favorable conditions for NPF to occur by releasing precursors and reducing the existing particle surface area (i.e., decreasing the condensation sink).

[...]

Overall, our results lead us to the hypothesis that meridional transport in combination with cloud processing and precipitation, as occurring in connection with, e.g., warm-air intrusions, may play an important role in free tropospheric Arctic NPF by delivering the ingredients (precursor gases including water vapor) to locations (high altitudes) with favorable conditions (intense solar radiation, low temperatures, small pre-existing particle surface area) for NPF.

13. LIne 346: In the ABL case study, figure 7c, The number concentration varied a factor of 2 within 30 minutes, a few times during the flight period. In figure 10c, the number concentration at cloud top is clearly actually the same as at cloud base at first, and goes up after a couple of scans. How can you be sure that the difference you see is actually because you are at cloud base or top, respectively, and not spatial/temporal variability?

Thank you for raising this point. The reviewer is right that we cannot definitely exclude that spatial and/or temporal variability contributed to the differences as well. Hence, we have included a sentence reflecting this uncertainty (lines 407 to 410):

For the comparison of the PNSDs measured at cloud top and cloud base, we only considered measurements carried out in the exact same geographical area, i.e., measurements between WP1 and WP2 from approximately 10:38 to 10:52 UTC (cloud base) and 11:17 to 11:31 UTC (cloud top). However, as differences are small and spatial and/or temporal variability cannot fully be ruled out, we cannot draw solid quantitative conclusions.

14. Figure 12: Why not show the no NFP cases like in Figure 5?

Thank you for this question. For the discussion on newly formed particles in the vicinity of clouds, we did not show the non-NFP cases as only about 15 minutes of consecutive measurements in cloud vicinity featured the absence of newly formed particles. Thus, any analysis with respect to these air masses is associated with large statistical uncertainties. In contrast, about 25 hours and 10 minutes of measurements were conducted in the free troposphere, of which 4 hours and 40 minutes indicated the presence of newly formed particles. In our view, for these cases, a substantive comparative analysis, discussing differences between NFP and non-NFP trajectories, can be carried out (Fig. 6 (Fig. 5 in the previous version)).
Therefore, we would like to stick to our original approach of not showing the non-NFP trajectories in Fig. 13 (Fig. 12 in the original manuscript).
No changes were made to the manuscript that relate to this point.

15. Line 390: Context missing, how much flight time over ocean and what would you expect during this season?

Thank you for this question. Please also see our response to the reviewer's comments #ii and #iii. The analysis of satellite-derived chlorophyll-a mass concentrations indicates that in the target regions which featured legs inside the ABL over completely ice-free ocean (on 11 and 25 April, in total about 120 minutes of measurements), no significant biological production (of precursors gases) took place. This might explain why no indications for the occurrence of newly formed particles over the completely ice-free ocean were found.
However, newly formed particles were found inside the ABL over ocean with sea ice floes. In this target region, the chlorophyll-a data suggest that a phytoplankton occurred.
We have revised Sect. 3.1 of the main text to summarize these results, and discuss the occurrence of newly formed particles inside the ABL over ocean with sea ice floes and the absence of newly formed particles inside the ABL over open water in detail in the SI in Sect. S5. Please find these revised sections in the response to the comments #ii and #iii. With respect to the statement mentioned here specifically, the revised version now reads (lines 448 to 451):
Specifically, these particles were detected in the free troposphere, inside the atmospheric boundary layer (ABL) over sea ice, in the vicinity of clouds, as well as inside the ABL over ocean with sea ice floes. No clear indications for newly formed particles inside the ABL over open, completely ice-free ocean were found.

16. Line 398: Where are the clear indications? I believe you have 2 arguments for this, one measurement of higher concentrations of small particles at cloud top (though not consistent throughout the measurement period) and a reference that cloud tops provide favourable conditions for NPF. I disagree that they constitute clear indications.

We thank the reviewer for the critical question. Although in our view the data presented here combined with previous results of NPF occurring directly

at cloud top suggest that this is a plausible interpretation, we agree that "clear indications" and "very likely" might be too strong at this point. We have therefore softened the corresponding formulation, which now reads (lines 460 to 462): Concerning newly formed particles observed at both cloud tops and cloud bases, we found indications that these particles are formed at the cloud tops and subsequently mixed down to the cloud bases, with the cloud tops providing a favorable environment for NPF to occur.
Please also see our response to the reviewer's comment #13, where we outline the changes made to manuscript discussing the potential contribution of spatiotemporal variability on our results.

17. Lines 404-405: I think the measurements you present, and the analysis of the 3 scenarios are unique and highly valuable to this research field. However, I think this statement is not really grounded in your data. There is no reason to believe that NPF in the Arctic FT is not geographically widespread, but these measurements also don't prove that they are. Also, what is geographically widespread? Be more precise, or less strong.

Thank you for this comment. Again, we have softened the statement and changed the sentence to (lines 466 to 469):
In particular, for the first time, our study presents observational results which indicate that NPF in the Arctic free troposphere occurred over distances of several hundreds of kilometers. In this context, a possible mechanism leading to Arctic free tropospheric NPF could be associated with warm-air intrusions.

18. The figures 5, 2d, 8, 11 and 12 are difficult to read when printed out because of the grid. Perhaps it can be made softer or sparser.

Thank you very much for this suggestion. We have decreased the linewidth of the plots' grids throughout the paper.

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