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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September 9, 2026

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

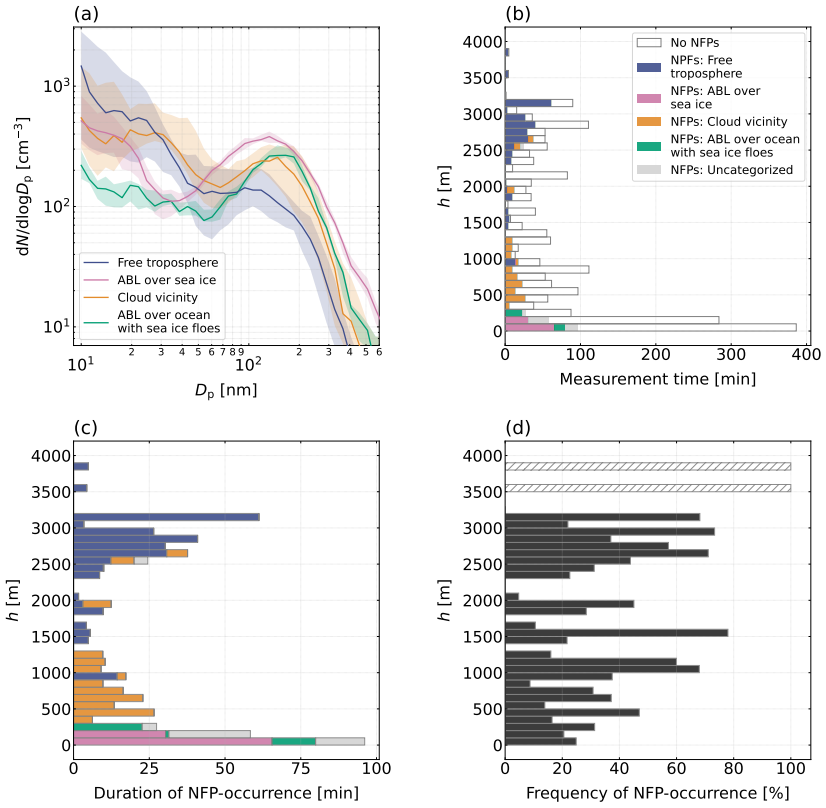


Figure R1: **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and 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

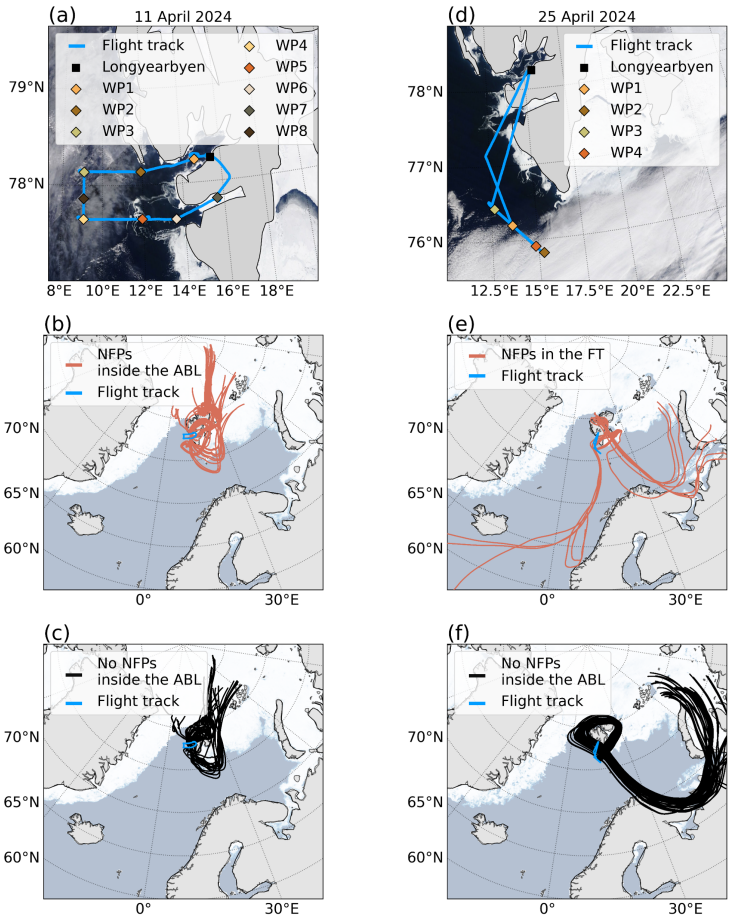


Figure R2: **(a)** Flight track of RF03 on 11 April 2024. **(b)** Five-day backward trajectories associated with newly formed particles (NFPs) inside the ABL on 11 April. **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles inside the ABL on 11 April. **(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).

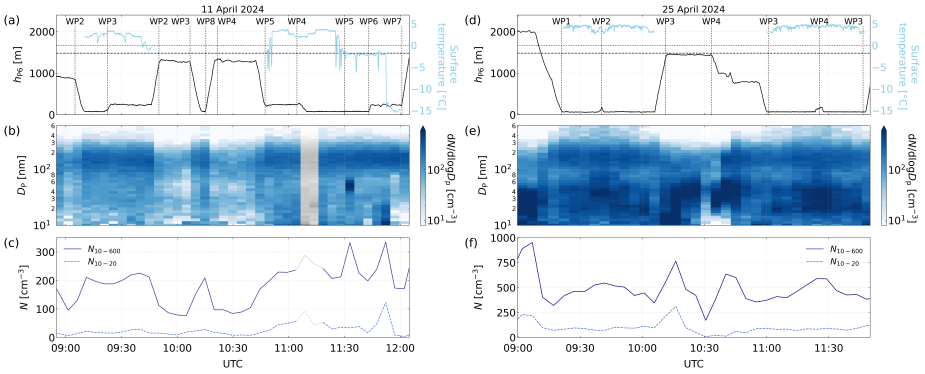


Figure R3: **(a)** P6 altitude during RF03 on 11 April 2024 (black line, left y-axis) as well as surface temperature (blue line, right y-axis). The dashed/dotted blue horizontal lines illustrate a surface temperature of 0°C and the freezing temperature of (salty) Arctic ocean water of −1.8°C. **(b)** 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.

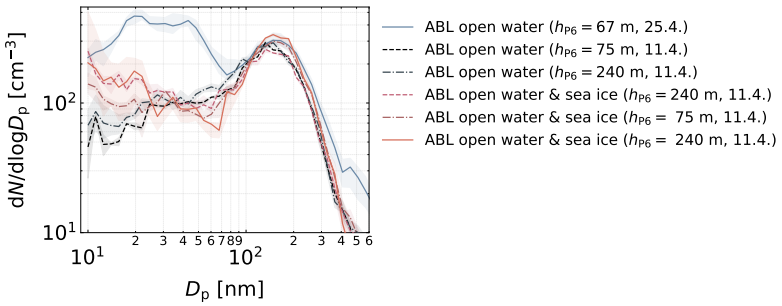


Figure R4: Median particle number size distributions during legs at constant altitude inside the ABL over completely ice-free open water (on 11 and 25 April, black and blue curves) and over ocean with both sea ice floes and open water areas present (on 11 April, red curves). The latter distributions 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).

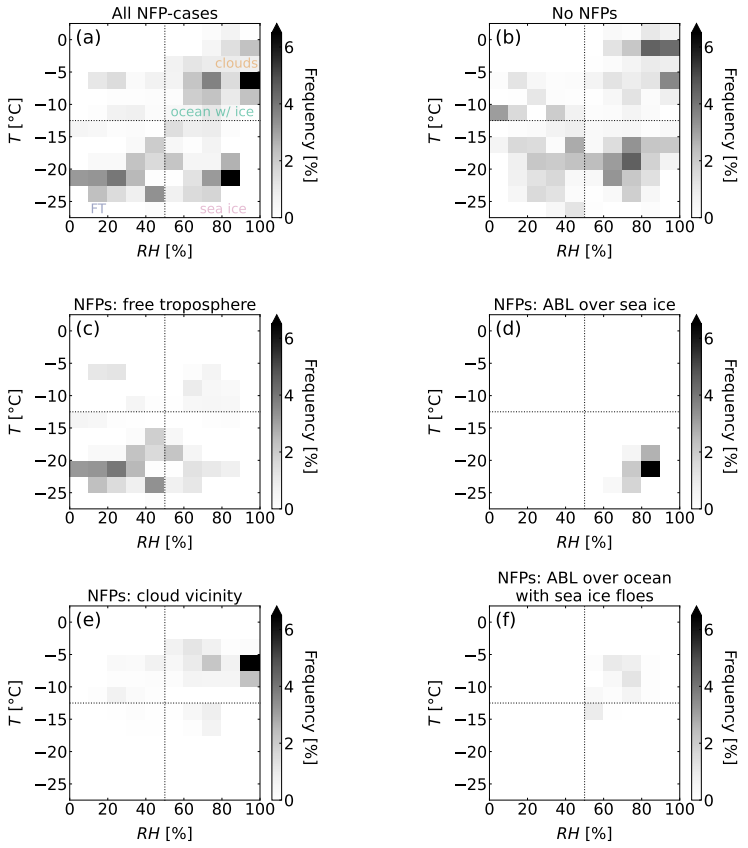


Figure R5: Temperature and relative humidity conditions during **(a)** newly formed particle-cases (NFP-cases) and **(b)** non-NFP-cases. The labels "FT", "sea ice", "clouds", and "ocean w/ ice" in panel (a) denote that within the corresponding quadrant, the measured temperature and relative humidity 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).

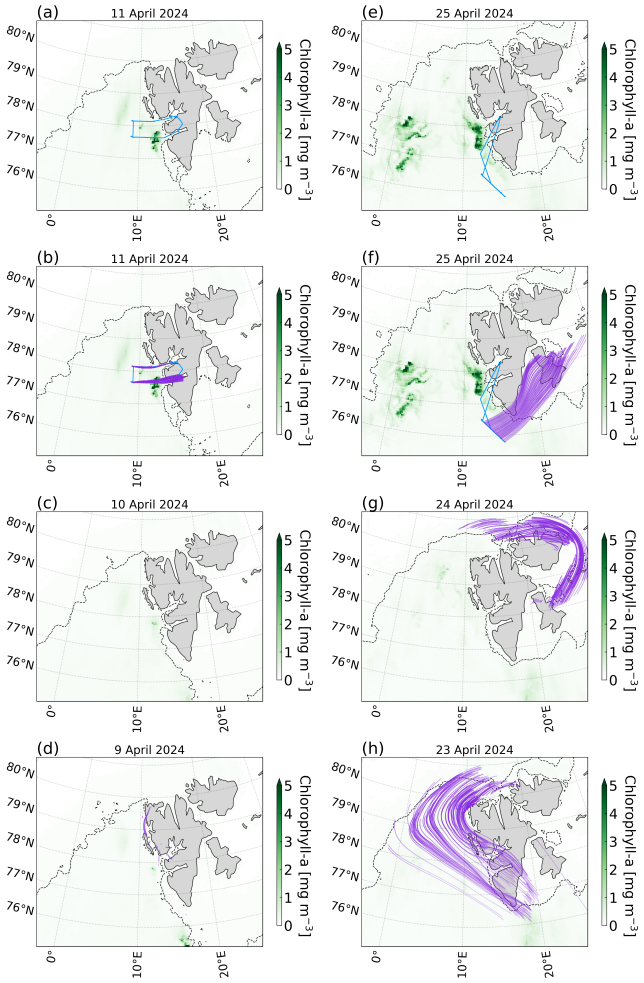


Figure R6: Flight track (blue), backward trajectories inside the ABL (violet), and chlorophyll-a mass concentrations (green) based on Copernicus-GlobColour data (Gohin et al. 2002; Hu et al. 2012) for the research flights featuring measurements inside the ABL over ocean on 11 and 25 April. The left column 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 $tas \approx 60\,\mathrm{m\,s^{-1}}$.

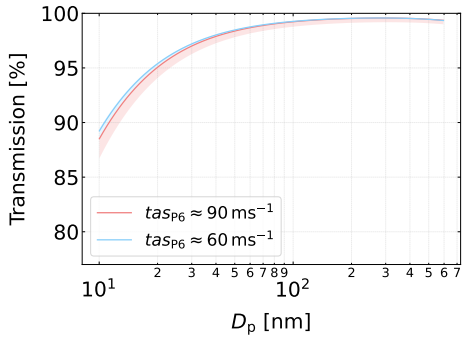


Figure R7: Particle size-dependent transmission efficiencies in the sampling tubing from the aerosol inlet to the MPSS for the two typical true airspeeds of the Polar 6 aircraft during the campaign.

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.

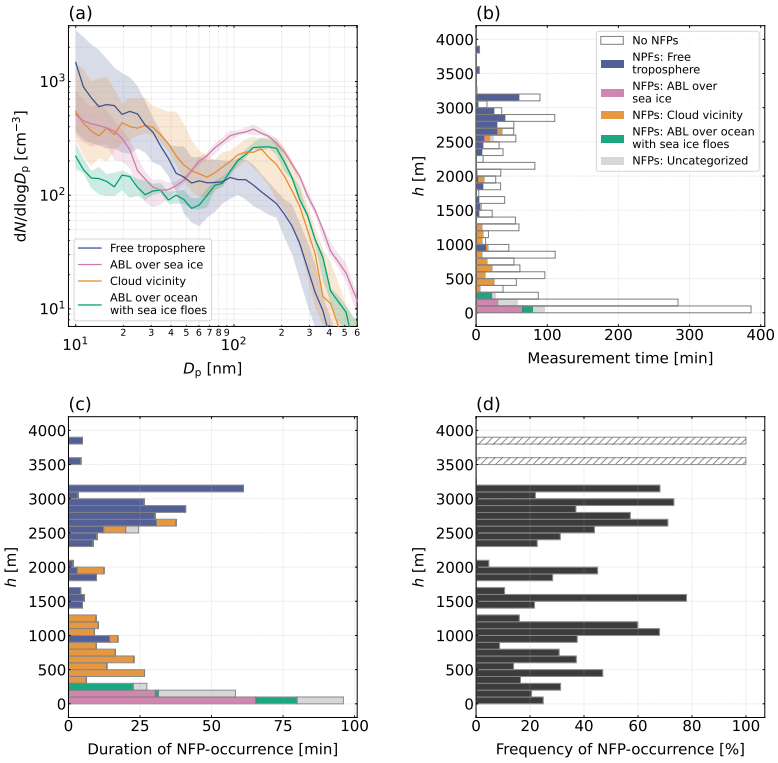


Figure R8: **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and 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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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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September 9, 2026

Summary

Reviewer #2: This study presents novel measurements of vertically-resolved aerosol size distributions in the Arctic, collected during a flight campaign that was carried out in the vicinity of Svalbard and Greenland in spring 2024. The measurements are further contextualised and investigated with the aid of Lagrangian back trajectories. The study focuses on the occurrence of newly-formed particles in the Arctic boundary layer and free troposphere, and as such constitutes a valuable contribution to a key research question regarding the role of the surface vs. the free troposphere in the sources and sinks of Arctic aerosol. I recommend that the study should be published after some minor revisions, detailed below.

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.

Main comments

Research gap and previous literature

i. the introduction and other parts of the manuscript rightly raise the question of boundary layer vs. free troposphere new particle formation as a key research gap in our understanding of Arctic aerosol, and highlight that vertically-resolved measurements of aerosol size distributions in the Arctic are currently lacking and are therefore of great value to the community. However, in my opinion there are certain lines in the manuscript that are either too strong or misrepresentative in their view of the conclusions of previous studies. For example, see lines 2-3 in the abstract ("Prior studies have predominantly investigated NPF in the Arctic atmospheric boundary layer (ABL), concluding that this phenomenon primarily takes place close to the surface."); line 49 ("These latter airborne studies predominantly suggested that NPF close to the surface inside the atmospheric boundary layer (ABL) prevails, and that free tropospheric NPF is less common."); or lines 190-196 ("The presence of newly formed particles in the Arctic free troposphere distinguishes our observations from previous airborne studies utilizing aircraft and helicopter measurements... these results led researchers to assume that NPF in the Arctic primarily occurs close to the surface").
It is true that many previous studies regarding observations of new particle formation/newly-formed particles in the Arctic have focused on surface measurements, but in my opinion, this is a result of the relative sparsity of vertically-resolved measurements, and not due to a particular consensus within the community that only the boundary layer is important, as is implied in these sections of the manuscript. I recommend revising these sections of the text to make this distinction clear.

We thank the reviewer very much for this helpful comment. We agree that some of our statements might have been formulated too strongly and could lead to misinterpretation. Still, specifically in lines 190-196 we referred to the following statements made in the publications by Willis et al. (2018) and Dall'Osto et al. (2017, 2020), which we cited in lines 195-196:

- "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)

Nevertheless, the reviewer is certainly right that no consensus in the community existed that only the atmospheric boundary layer (ABL) was an important source of newly formed particles, and that the sparsity of vertical measurements, in particular compared to many ground-based studies, has led to the majority of studies on Arctic NPF discussing events taking place inside the ABL. We are grateful to the reviewer's comment and have reformulated the abstract, the introduction, and the discussion for the case of newly formed particles in the free troposphere in the results section. Please find the revised versions 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 impor-

tance 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 295):
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 helicopter (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.

ii. I also note on this subject the Community Comment posted on 29 May 2026 relating to another previous study that presented measurements of newly-formed particles in the Arctic free troposphere. The authors should consider how that study relates to the discussion within this manuscript and include as needed.

The study we were made aware of by the Community Comment is indeed highly relevant to our present manuscript. We have included a corresponding reference and respective discussions in the introduction and the discussion on newly formed particles detected in the free troposphere, as outlined in our response to the reviewer's comment #i. Please find these revised sections above.

Classification of NFP periods

iii. it is not overall clear how the three NFP regimes (free troposphere, ABL above sea ice, vicinity of clouds) were selected. Are these simply individual case studies that were selected by the authors? The fraction of "unclassified" NFP observations is fairly high (figure 2a), almost as high as ABL over sea ice. Figure 2b puts them mostly in the ABL. What kind of conditions do they correspond to?

Thank you for raising this point! These regimes correspond to the occurrence of newly formed particles which could clearly be assigned to specific environmental conditions based on our in situ data. Naturally, because a flight campaign with a somewhat limited spatial and temporal coverage cannot deliver representative statistics/measurements for a certain Arctic region, these scenarios are influenced by the specific scientific questions and regions targeted during the campaign. For example, both the ABL in sea ice-covered areas and the tops and bases of Arctic clouds were of specific interest to us during our campaign, as well as probing air masses transported to the Arctic from mid-latitudes within warm-air intrusions. When analyzing the particle number size distribution (PNSD) data collected throughout the campaign, we noticed that about 86 % of PNSDs featuring newly formed particles were associated with one of the three regimes described in the originally submitted manuscript. Nevertheless, as noted by the reviewer in the next comment #iv, these different scenarios are not intended to describe an exhaustive framework for the occurrence of newly formed particles.

Motivated by the reviewer's comment, we have carried out a more detailed analysis of the unclassified cases. 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. These cases are now included as a separate 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.

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 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 ≈ 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. R1b, c). 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. 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

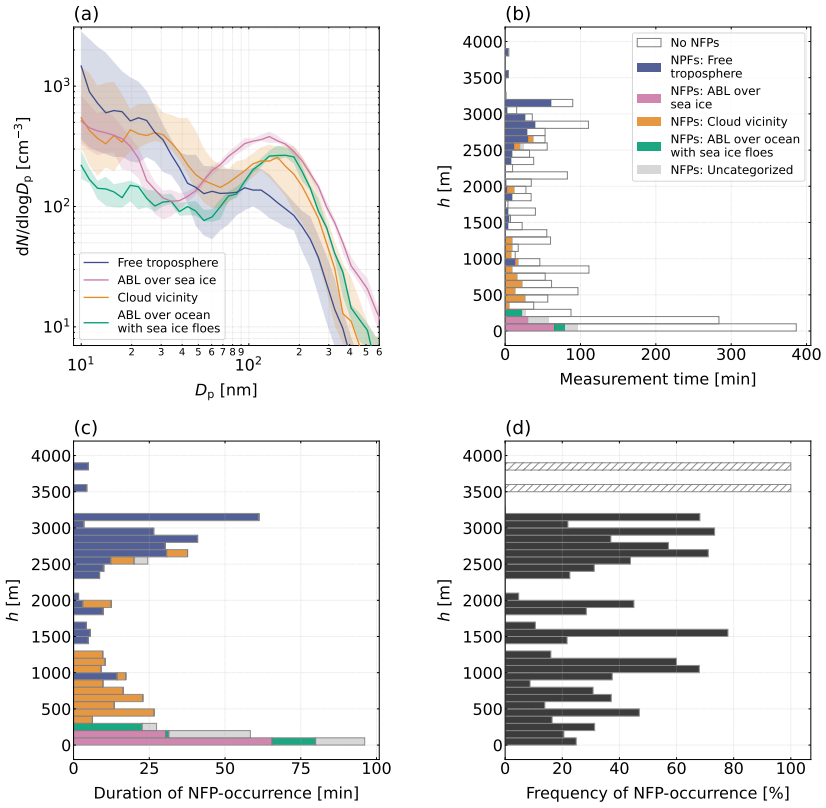


Figure R1: **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and 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).

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 - 210):

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 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

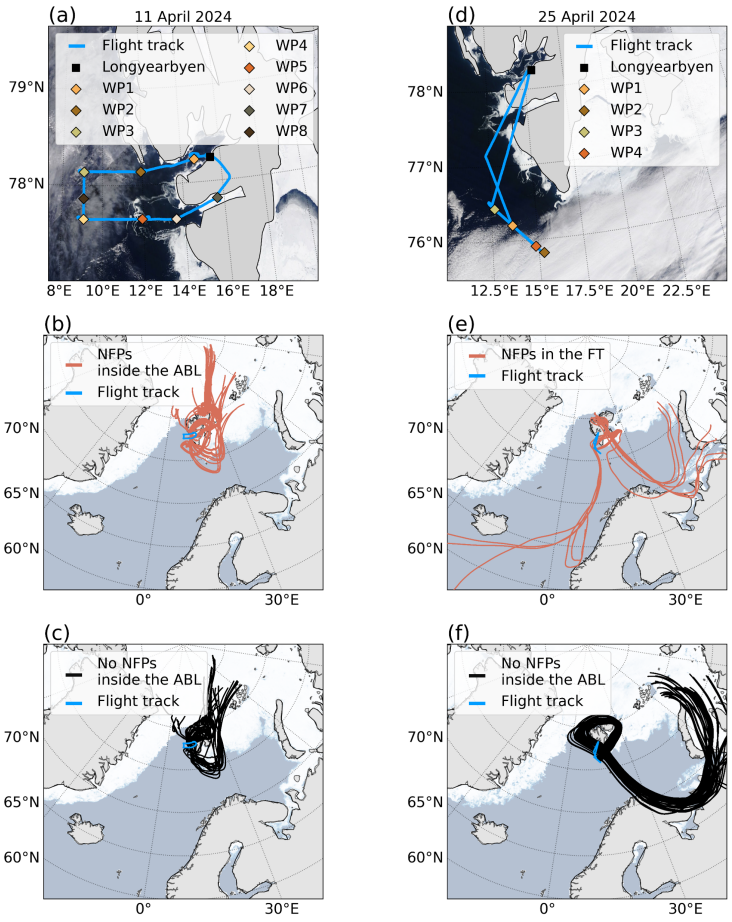


Figure R2: **(a)** Flight track of RF03 on 11 April 2024. **(b)** Five-day backward trajectories associated with newly formed particles (NFPs) inside the ABL on 11 April. **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles inside the ABL on 11 April. **(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).

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).

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 ≈ 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

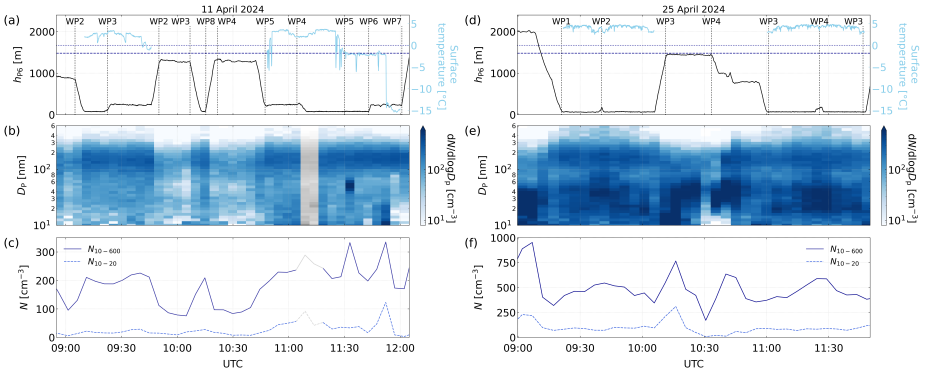


Figure R3: **(a)** P6 altitude during RF03 on 11 April 2024 (black line, left y-axis) as well as surface temperature (blue line, right y-axis). The dashed/dotted blue horizontal lines illustrate a surface temperature of 0°C and the freezing temperature of (salty) Arctic ocean water of −1.8°C. **(b)** 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).

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.

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

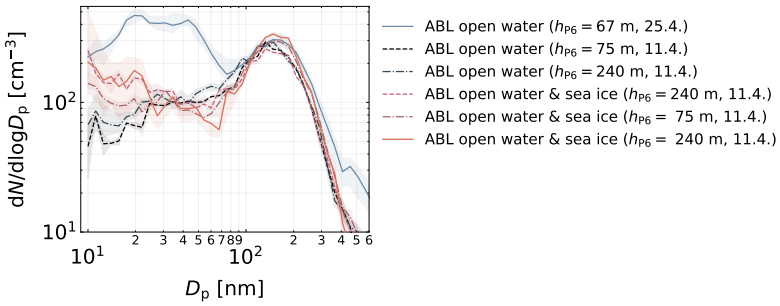


Figure R4: Median particle number size distributions during legs at constant altitude inside the ABL over completely ice-free open water (on 11 and 25 April, black and blue curves) and over ocean with both sea ice floes and open water areas present (on 11 April, red curves). The latter distributions 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.

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. R5a) 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. R5a).

Fig. R5b 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. R5b-d, f-h. As mentioned above, easterly winds prevailed in both the northern and the southern section (Fig. R5b). 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. R5b). 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

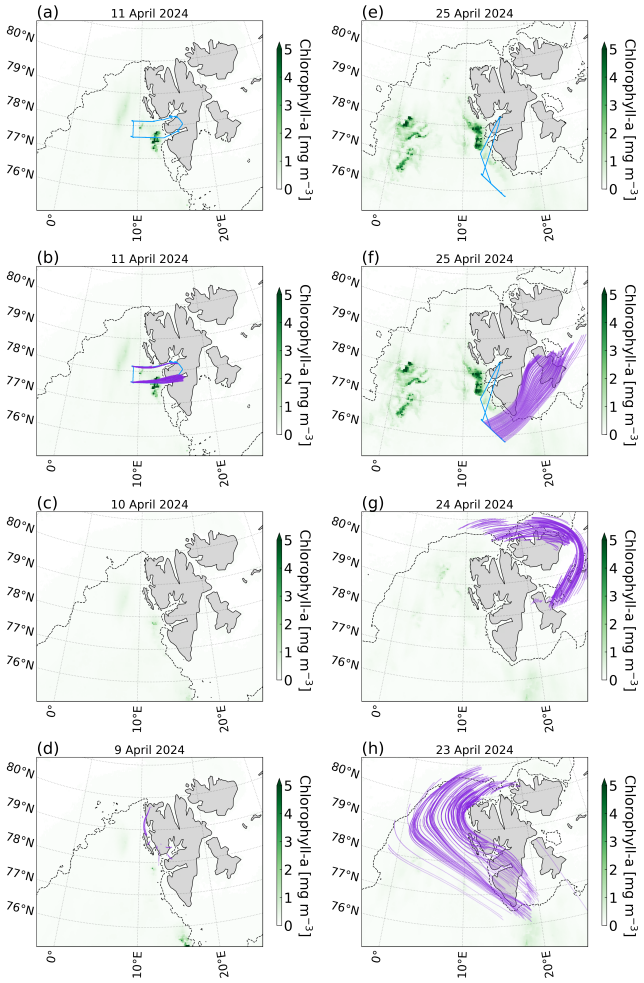


Figure R5: Flight track (blue), backward trajectories inside the ABL (violet), and chlorophyll-a mass concentrations (green) based on Copernicus-GlobColour data (Gohin et al. 2002; Hu et al. 2012) for the research flights featuring measurements inside the ABL over ocean on 11 and 25 April. The left column 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).

bloom. On the two previous days, the air massed resided completely (10 April, Fig. R5c) and overwhelmingly outside of the ABL (9 April, Fig. R5d), 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. R5e). 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. R5f-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. R5b). 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).

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. R6).

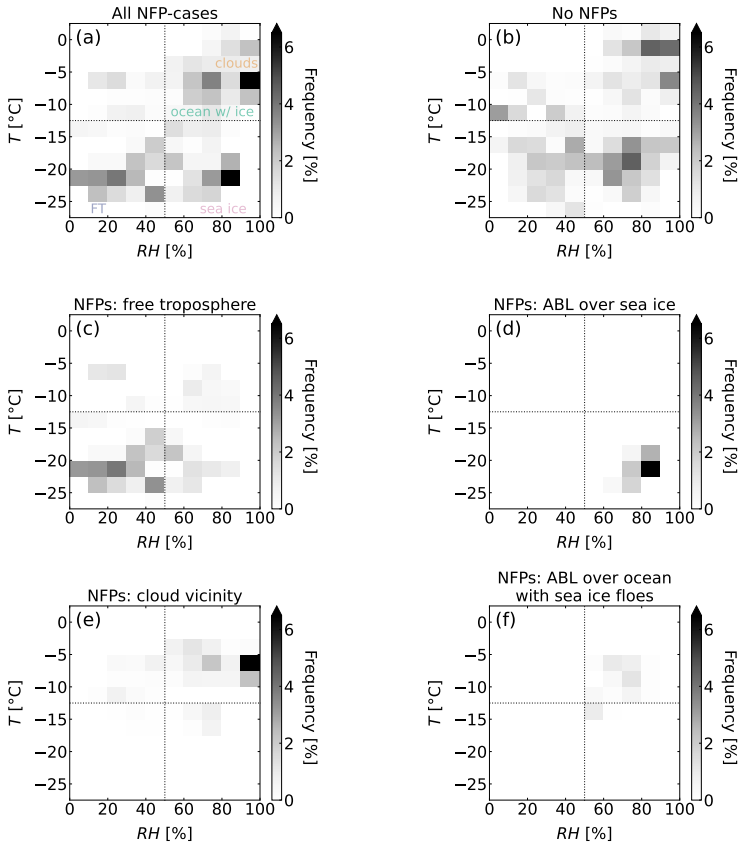


Figure R6: Temperature and relative humidity conditions during **(a)** newly formed particle-cases (NFP-cases) and **(b)** non-NFP-cases. The labels "FT", "sea ice", "clouds", and "ocean w/ ice" in panel (a) denote that within the corresponding quadrant, the measured temperature and relative humidity 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.

Furthermore, we have included an additional section in the SI to explain to the reader the difficulties associated with determining the specific environmental conditions precisely for all flight section throughout the campaign, which is the reason why some unclassified cases of newly formed particle-occurrence remain. Please find the section below (lines 32 to 64 in the SI):

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, approximately 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.

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.

iv. Similarly at line 388, ("Specifically, these particles were detected in the free troposphere, inside the atmospheric boundary layer (ABL) over sea ice, and in the vicinity of clouds. No clear indications for newly formed particles inside the ABL over open ocean were found.") it is implied that these three regimes describe an exhaustive framework for all conditions under which NFPs were observed. I would recommend a more in-depth discussion of the unclassified events and how the three regimes were selected to support this conclusion more strongly, or otherwise to amend this text.

Thank you very much for this recommendation! We have included a discussion on the observations inside the ABL over ocean, on the unclassified events (and found another fourth regime), as well as a description on how the (now) four regimes were selected. In this regard, please also see our response to reviewer's comment #iii. We have revised section 3.1 of the main text and included additional sections in the SI (Sect. S3, Sect. S5). Please find these revised sections in the response above. Specifically with respect to the statement mentioned by the reviewer here, which is placed in the conclusion section, 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.

Conclusions about process-level drivers and broader relevance of case studies

v. in my opinion, some of the conclusions made for each of the three regimes are put too strongly compared to what the data shows. Although many of the hypotheses that the authors put forward can certainly be argued to be consistent with what is presented, I do not agree in every case that the data here is enough to rule out other possibilities definitively. Specifically:

Line 273: "The spatial variability of the number concentration of nucleation mode particles suggests geographically heterogeneous processes and points toward newly formed particles as the result of regional formation rather than long-range transport, as the latter would likely lead to rather constant concentrations of newly formed particles along the flight track."
And 280: "This temporal variability points toward a regional source as well, since it suggests that we observed an NPF event that becomes weaker throughout the day"
I struggle to understand this argument that spatial/temporal heterogeneity in number concentration rules out long-range transport. What are the mechanisms that would remove such variability in the nucleation mode if it was formed in an air mass that has been transported? I would be grateful if the authors could explain this argument further.

Thank you for this question! In general, short atmospheric residence times of nucleation mode particles due to diffusion, coagulation with larger particles, and condensational growth out of the nucleation mode size range yield long-range transport of newly formed particles rather unlikely (typical residence times of $\lesssim 1$ day for ~ 10 nm particles; e.g. Wallace and Hobbs (2006) and Pruppacher and Klett (2010)). These process tend to smear out concentration gradients of newly formed particles particles. During the case study on newly formed particles inside the ABL over sea ice, we, however, observed rather strong spatial as well as temporal differences of nucleation mode particles (N_{10-20}). In Fig. 8 (Fig. 7 in the previous version), the number concentration of N_{10-20} varies by a factor of roughly 10 between maximum and minimum concentrations during the low-level legs inside the ABL. For Arctic measurements of newly formed particles, Kupiszewski et al. (2013) argued similarly when allocating sharp increases of nucleation mode particles to local particle production instead of long-range transport.

In our view, it appears feasible to deduce that this rather indicates a local or regional source. Nevertheless, already in the original manuscript, we had mentioned that "we are not able to unambiguously resolve if the precursors involved in NPF originated directly from the sea ice-covered measurement region, with various cracks and leads being present (Fig. S6), or if the precursors were transported to the target area from the higher Arctic". Similarly, we noted that "we hypothesize that the observed presence of newly formed particles in the ABL over sea ice is connected to precursor gases (conceivably iodine components) which originate from natural, most likely regional, but possibly also higher-latitude Arctic sources within the sea ice, from leads, or snow." We thus believe that we have already discussed that we cannot ultimately prove that regional NPF is in fact what occurred here, and that we have addressed the corresponding uncertainties. Still, to strengthen our argument, we have modified the respective statement, which now reads (lines 330 to 335):

The spatial variability of the number concentration of nucleation mode particles points toward newly formed particles as the result of regional formation rather than long-range transport. Nucleation mode particles have short atmospheric residence times due to their high diffusion, coagulation with larger particles, and condensational growth out of the nucleation mode size (e.g., Wallace and Hobbs 2006; Pruppacher and Klett 2010). These effects tend to smooth out concentration gradients of such small particles, thereby making a contribution from long-range transport to the observed events less likely (Kupiszewski et al. 2013).

vi. Line 394: "With respect to the newly formed particles detected inside the ABL over sea ice, we conclude that these particles are presumably the result of regional NPF events induced by precursor gases (conceivably iodine components) originating from the sea ice, leads, or snow present in the measurement region." Related to the point above, I believe the measurements presented under section 3.3 could indeed be consistent with a source related to precursors from the sea ice, leads, or snow, but I do not feel that the results in section 3.3 prove

this robustly.

We thank the reviewer for raising this point. We agree that without further measurement data, e.g., of precursor gases, we cannot prove definitely that the precursors are emitted from the sea ice, leads, or snow. Nevertheless, we are convinced that our data present indications in support of the hypothesis about regionally sourced newly formed particles, as outlined in the response above to the reviewer's comment #v. Already in the original manuscript, we had relativized our statement by noting that "contributions of higher-latitude Arctic (precursor) sources cannot be ruled out". Still, to further soften the corresponding sentence, we have replaced "conclude" by "hypothesize" (lines 457 to 460):
With respect to the newly formed particles detected inside the ABL over sea ice, we hypothesize that these particles are the result of regional NPF events induced by precursor gases (conceivably iodine components) originating from the sea ice, leads, or snow present in the measurement region. However, contributions of higher-latitude Arctic (precursor) sources cannot be ruled out.

vii. Line 397: "Concerning newly formed particles observed at both cloud tops and cloud bases, we found clear indications that these particles are very likely 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." If I understood correctly, in section 3.4 it is argued that because the number concentrations of the nucleation and Aitken modes are higher at cloud top than at cloud base, this indicates that formation occurs at cloud top followed by growth within cloud that causes fewer particles to be observed in these sizes at cloud base. While this interpretation is believable, I think that the words "clear indication" and "very likely" are too strong here without further evidence that this is what takes place (e.g. dedicated large-eddy simulations of the event).

Agreed. We have changed the sentence to (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.

Furthermore, we have included a sentence addressing that spatial or temporal variability might be a contributing factor to the differences between PNSDs measured at cloud top and cloud base (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.

viii. Line 206: "Our data support these latter observational results and

indicate that NPF might occur frequently in the Arctic free troposphere over large geographical areas." The words "frequently" and "large geographical areas" here are somewhat subjective, I recommend to be more precise. While the measurements in this study are indeed novel and valuable, the frequency or geographical extent of the processes discussed here cannot be assumed outside of this field campaign.

We thank the reviewer for raising this valid point. We have replaced the respective sentence with the following statement (lines 294 to 295):
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.
Please find the full revised paragraph above in the response to the reviewer's comment #i (lines 276 to 295 in Sect. 3.2).

ix. Line 403: "In particular, for the first time, our study presents observational results which indicate that NPF in the Arctic free troposphere could be a geographically widespread phenomenon, and that free tropospheric NPF is connected to warm-air intrusions." Here I raise the same point as above about the use of "geographically widespread". Also, though the discussion about warm air intrusions based on the results in figure 5 is very interesting and certainly warrants further attention, I would recommend to soften the phrase "free troposphere NPF is connected to warm-air intrusions". The data presented here does not rule out, for example, that free tropospheric NPF could also take place outside of warm air intrusion conditions.

Thank you for this comment. We agree with the reviewer and have softened and reformulated the corresponding statement 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.

Minor comments

1. Figure 3b (also to lesser extent figure 6b and 9b): I find it somewhat difficult to distinguish non-NFP trajectories from NFP trajectories. Please consider separating into two subplots or adjusting the transparency of the NFP trajectories.

Thank you very much for this suggestion which clearly helps to differentiate between NFP and non-NFP trajectories! We have separated the NFP and non-NFP trajectories into two suplots and modified Fig. 3 (Fig. 4 in the revised manuscript), Fig. 6 (now Fig. 7), and Fig. 9 (now Fig. 10) accordingly.

2. Figure 5 vs figure 8: Could the two sets of trajectories in figure 8 be combined into one average NFP trajectory for consistency with figures 5 and 12? From what I understand of the text in section 3.3, the only key difference between the results of the two trajectories is that there was more contact with clouds in the trajectories arriving on 15th April than for those on 13th April. This could perhaps be summarised in the text and/or with a supplementary figure rather than including both sets of trajectories separately in the main figure and describing them in the main text.

Thank you for this suggestion. Following the argument of the reviewer, we have combined the two sets of trajectories into one average trajectory and mentioned the only key difference (the reviewer is right here) regarding the last cloud contact in the corresponding section of the main text (lines 357 to 360): For about 60 % of the trajectories, the last cloud contact occurred approximately 24 h before arrival in the target region (Fig. 9j). Roughly 15 % of the trajectories were not influenced by clouds on their way to the measurement location during the last five days, including 80 % of all trajectories on 13 April.

3. Figures 5, 8 and 12 (also see comment above): although figure 5 aids the comparison of NFP vs. non-NFP conditions for the first case of free troposphere conditions, the equivalent two figures for the other two cases do not provide this direct comparison, presumably because the non-NFP periods are not available in the other two cases. As such, perhaps it would be more informative to combine figures 5, 8 and 12 so that the NFP trajectories of each case can be directly compared. The non-NFP trajectories of the free troposphere case could also be included.

We thank the reviewer for this remark. Indeed, for the other two cases, the non-NFP periods are not available in any meaningful way. That is, only about 15 minutes of consecutive measurements in cloud vicinity featured the absence of newly formed particles. Similarly, for only about 20 minutes of measurements inside the cloud-free ABL over sea ice, no newly formed particles were detected. 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. Here, in our view, a meaningful comparative analysis, discussing differences between NFP and non-NFP trajectories, can be carried out (Fig. 6 (Fig. 5 in the previous version)). Further, we believe that the differences between NFP and non-NPF trajectories in the free troposphere comprise essential results, needed to explain the observations of free tropospheric newly formed particles. In our view, it is therefore important to discuss the differences in the corresponding subsection (Sect. 3.2), in proximity to the respective case study on newly formed particles. Thus, we would like to stick to our original structure and approach of showing the trajectory analysis for the two other cases in the respective sections as in the submitted manuscript, for the sake of consistency.

No changes were made to the manuscript which relate to this point.

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

David J. Simon , Jonas Schaefer, Jörg Hartmann, Benjamin Kirbus, Joshua Müller, Markus Hartmann, Bruno Wetzel , Laura Köhler, Anna-Marie Jörss, Kay Weinhold, Andreas Herber, Zsófia Jurányi, Silvia Henning, Gregory C. Roberts, Manfred Wendisch, Mira L. Pöhlker, and Frank Stratmann

September 9, 2026

Farahnaz Khosrawi: I would like to raise the attention of the authors to the following study which I published a while ago on new particle formation in the Arctic free troposphere:

Khosrawi, F., Ström, J., Minikin, A., and Krejci, R.: Particle formation in the Arctic free troposphere during the ASTAR 2004 campaign: a case study on the influence of vertical motion on the binary homogeneous nucleation of H2SO4/H2O, Atmos. Chem. Phys., 10, 1105–1120, https://doi.org/10.5194/acp-10-1105-2010, 2010.

It would be worth mentioning this study in the introduction on e.g. P2, L50 and/or the paragraph below. Our study analysed measurements from the ASTAR 2004 campaign and frequent newly formed particles in the free troposphere up to 4000 m were there also reported.

On P6, L135 and/or 157: The Khosrawi et al. (2010) study may be mentioned here as well.

P9, L197: Here, it would be definitely worth to mention here the Khosrawi et al. (2010) study where we investigated a case where newly formed particles were observed at 7000 m.

Author reply: Thank you very much for pointing us to your interesting study! Indeed, we were unfortunately not aware of your paper, but believe it is highly relevant to our present study. Please find our detailed response below. Your comments are marked in black, our respective answers in green, and resulting changes in the manuscript and supporting information in blue.

Regarding your study (Khosrawi et al. 2010), we have included references and discussions at appropriate positions. Specifically, we have modified the following paragraphs:

Introduction (lines 49 to 57 in the revised manuscript)

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. A number of studies reported 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).

Results and Discussion

Section 3.2 (Newly formed particles in the free troposphere, lines 276 to 295)

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 helicopter (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.

Farahnaz Khosrawi: P3, L84: The distinction of NPF and newly formed particles is here a bit weird since reading the sentence it feels like the same. You later explain the difference and then it becomes clear. My suggestion would be to move this sentence after explaining the difference between NPF and newly formed particles.

Thank you very much for this suggestion, which has helped us to improve the readability and clarity in this section! We have slightly modified the order of the sentences in this paragraph which now reads (lines 93 to 98):
Since the minimum diameter of aerosol particles that can be detected with our measurement instrumentation is 10 nm, it is not feasible to clearly identify the exact region where NPF occurred, i.e., where the process of the initial nucleation of particles with sizes of about 2 to 3 nm from condensable gases took place (Kulmala et al. 2004; Kerminen et al. 2018). Nevertheless, particle sizes of 10 nm still imply that these particles were newly formed by secondary particle formation (i.e., by NPF, Kulmala et al. 2004). We hence refer to these particles as newly formed particles and, in this paper, instead on the process of NPF, focus on the occurrence of such secondarily formed particles.

Farahnaz Khosrawi: P14, L314: Is descent here really correct? If precursors were emitted from the surface, I would expect that these are then transported upwards, thus that you observe an ascent of air masses.

Thank you for this question. Descent is in fact correct at this point. The descent of the air mass into the atmospheric boundary layer (ABL) provides the possibility that this air mass takes up precursors gases (from the surface). If the air mass had not descended in to the ABL but remained in the free troposphere, possibly separated by a strong temperature inversion as common over the Arctic pack ice, mixing between the surface and the air mass would have been considerably limited. This would have very likely prevented precursor gases from the surface to be emitted in to the air mass. No changes were made to the manuscript which relate to this point.

Farahnaz Khosrawi: P14, L316-318: Isn' that a bit too speculative? Are there any measurements or other studies that could support this hypothesis?

You are of course right that, due to missing trace gas measurements, we cannot ultimately prove that precursor gases were emitted in the measurement region. Nevertheless, in our view, our measurements support the hypothesis of

a regional source, mostly due to the spatial and temporal heterogeneity of nucleation mode particles, but also because the trajectory analysis points toward this direction as well, as outlined in these paragraphs in Sect. 3.3 you referred to. In order to clarify that already previous studies have found newly formed particles in sea ice-covered regions in the Arctic and connected those observations to iodine components, we have included references to the respective studies here again. Please find the revised sentence below (lines 373 to 376):
In summary, from our observations, we hypothesize that the observed presence of newly formed particles in the ABL over sea ice is connected to precursor gases (conceivably iodine components, as reported by Allan et al. 2015; Sipilä et al. 2016; Baccarini et al. 2020; L. J. Beck et al. 2021) which originate from natural, most likely regional, but possibly also higher-latitude Arctic sources within the sea ice, from leads, or snow.

Farahnaz Khosrawi: P18, 382: Add "free" so that it reads "Arctic free troposphere"?

Author reply: Thank you for this question. In fact, we here refer to our measurements covering a large range of altitudes in the lower troposphere, i.e., both the atmospheric boundary layer (ABL) as well as the free troposphere. The "free" is therefore omitted on purpose. No changes were made which relate to this point.

Farahnaz Khosrawi: P20, L405: I am not convinced of this statement. Why do you think that NPF could be a geographically widespread phenomen? How do you see that in your data/analysis? Aren't you focusing on a certain part of the Arctic (a certain distance around the flights that are only a minor part of the entire Arctic). However, why shouldn't NPF occur everywhere in the Arctic? There are so many possibilities to form new particles so that I would assume that NPF can occur nearly everywhere in the Arctic. Further, how you show the connection to warm air intrusions would be worth to be discussed/repeated here a bit more.

Author reply: Thank you very much for this comment! Following your feedback as well as the comments by the the two referees, we have toned down this statement. The corresponding lines now read (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.

Farahnaz Khosrawi: P20, L400ff: Are there already any studies or measurements that investigate the trends or changes in NPF due to climate change?

For example, Dall'Osto et al. (2017, 2018) observed a correlation between an increase of NPF events and decreasing sea ice extent (which occurs due to

climate change). Dada et al. (2022) reported on an extreme aerosol event in the central Arctic, which was triggered by a warm-air intrusion. This transport of warm and moist air from lower latitudes is expected to occur more frequently in the future (Bintanja et al. 2020; Wendisch et al. 2025), which might possibly also affect the occurrence of (free tropospheric) NPF in the Arctic, as discussed in our paper. 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). These previous studies are all mentioned in the introduction, where the latter sentence referencing Heslin-Rees et al. (2020) has been included in the revised version of the manuscript following the comment of reviewer #1. As we cannot contribute any quantitative insights with respect to possible changes of NPF due to climate change based on a single aircraft campaign, but studies on long-term ground-based measurements are much more suited to do so, we believe that this aspect is not an essential part of our manuscript. Therefore, in our view, the previous literature is already discussed sufficiently in the introduction and we would like to keep the outlook rather concise at this point. No further changes in addition to the inclusion of the Heslin-Rees et al. (2020) study were made which relate to this comment.

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)."

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September 9, 2026

Caroline Leck: Although the manuscript cites many highly relevant articles from the field, the literature survey is incomplete; substantial prior observational evidence appears to have an essential bearing on the results and conclusions and thus merits discussion. In this respect, it seems beneficial to mention or learn from the previous work by Tjernström, Heintzenberg, Leck, and their colleagues over the past more than three decades over the Arctic pack ice area (including the marginal ice zone) and at the Mt. Zeppelin Observatory (Spitsbergen).
Their work has shown that it is not only necessary to specify particle concentrations in the atmosphere and their possible sources, but we must also understand the thermodynamic structure of the lower atmosphere (typically a well-mixed, shallow boundary layer at the surface, only a couple hundred meters deep, capped by a temperature inversion below the free troposphere), the dynamics of the boundary layer, and processes important for exchange between the air and the ocean's top layers.

Author reply: Thank you very much for your comment and for providing a detailed summary on prior work by Tjernström, Heintzenberg, Leck, and colleagues. We appreciate your feedback which has helped us to improve our manuscript. Please find a detailed response below. Your comments are marked in black, our replies in green, and corresponding changes in the manuscript in blue.

1. Over the pack ice, the structure of the lower atmosphere generally limits mixing of aerosol particles in the free troposphere into the boundary layer. For this to happen, aerosol sources in the free troposphere (either locally sourced or advected from lower latitudes) must be brought down to the top of the boundary layer, where entrainment can occur. The only mechanism that can bring elevated plumes down to the inversion is large-scale subsidence, which is a very slow process. Entrainment is thus unlikely to be a major factor in CCN (Aitken/accumulation modes) number concentrations within the lower atmosphere and will not be a main contributor to the formation of low-level clouds (e.g., Bigg et al., 2001; Tjernström et al., 2012).

We thank you very much for this comment. It is of course correct that the stratification of the atmosphere will significantly influence the mixing between the free troposphere and the ABL and thus if, e.g., low-level clouds can be affected by free tropospheric particle sources. We are grateful for your remark and have included the following discussion in the section on newly formed particles in the free troposphere.
Section 3.2 (lines 296 to 303 in the revised manuscript)
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.

2. Also not accounted for is the possibility that aerosols and their precursors, lofted in the deeper atmospheric upstream well mixed boundary layer over the open water, could be advected in over the pack ice on top of the shallow local boundary layer (Tjernström et al., 2012) and later be entrained into the local boundary layer through the cloud top by cloud induced mixing (Igel et at., 2017; Brooks et al., 2017; Shup et al., 2013). On this note, Wiedensohler et al. (1996) concluded that the ultrafine mode was produced by nucleation in the upper layers of the marine boundary layer and mixed down to the measuring site, ca. 25 m asl.

Thank you for pointing us to these studies! Please find the main changes made to the manuscript in the response to your comment #2 above. Here, Shupe et al. (2013) was already mentioned in the discussion on newly formed particles in the vicinity of clouds. Following your comment, we additionally

included a reference to Wiedensohler et al. (1996) in the introduction (lines 40 to 43):
NPF has been observed in several locations Arctic-wide (Willis et al. 2018; Schmale and Baccarini 2021). Most studies were based on measurements conducted during shipborne campaigns (e.g., Wiedensohler et al. 1996; Allan et al. 2015; Baccarini et al. 2020) or at ground-based stations including Alert, Villum, Zeppelin, Tiksi, and Utqiagvik (e.g., Leaitch et al. 2013; Asmi et al. 2016; Croft et al. 2016; Freud et al. 2017; Beck et al. 2021).

3. Another past observation over the pack ice that merits discussion is the demonstration that organics found in Aitken- and accumulation-mode aerosols and in cloud water behaved like marine polymer gels originating from the surface microlayer on leads (open water between ice floes), due to the activity of ice microalgae, phytoplankton, and perhaps bacteria (e.g., Leck and Bigg, 2005; Leck et al., 2025; Bigg and Leck, 2008; Orellana et al., 2011; Leck et al., 2013; Hamacher-Barth et al., 2016).

Thank you for pointing us to these interesting studies. Indeed, the breakdown of marine polymer gels offers a possible explanation for the occurrence of particles in the nucleation mode size range, in particular for the case of the occurrence of such particles inside the ABL over ocean with sea ice floes on 11 April, with the sea surface containing both open water as well as sea ice. We have included this hypothesis into our manuscript into Sect. S5 (lines 202 to 210):
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 proposed initially 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).

4. Another essential factor affecting the conclusions is that elevated particle number concentrations have been observed despite markedly different regional meteorological conditions and have persisted for several hours to a day. These consist of particles approximately 10 nm across, those in the 30–50 nm range, and smaller particles (2.5–5 nm in diameter) that can reach concentrations of up to about 1000 particles per cm^3 (Karl et al., 2013; Lawler et al., 2021; Leck and Bigg, 1999; 2010; Leck et al., 2025). Their presence does not rely on a source from the free troposphere.
Such episodes often involve rapid droplet dissipation from the evaporation of

intermittent fogs or low cloud bases/tops, coinciding with the peak periods of sub-5 nm particle enhancements. The formation of aerosols at the condensation-evaporation interface, especially within the sub-Aitken mode size range, is thus well established.

Thank you for this comment and for elaborating on these interesting publications! We have included a reference regarding the formation of aerosol particles linked to dissipation of cloud or fog droplets in our manuscript. Please find the changes made to the manuscript in our response to the previous comment #3 above.
However, we respectfully disagree that these observations affect our conclusions. For example, we did not state that the free troposphere was the only important source of particles and that elevated particle number concentrations could not be present without free tropospheric sources. On the contrary, our observations highlight both the importance of free tropospheric as well as sources within the ABL. No further changes expect for those outlined above were made to the manuscript which relate to this comment.

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Frequent occurrence of newly formed aerosol particles over wide geographical areas in the Arctic free troposphere and atmospheric boundary layer

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Abstract. 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 ~~studies~~ measurements have predominantly investigated NPF in the Arctic atmospheric boundary layer (ABL), ~~concluding that this phenomenon primarily takes place close to the surface. However, this study shows~~ 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 three~~, and identified four atmospheric scenarios for their occurrence: newly formed particles in the free troposphere, in the ABL over sea ice, ~~and~~ in the vicinity of clouds, and in the ABL over ocean with sea ice floes. Our results ~~suggest~~ indicate that regional Arctic atmospheric processes as well as long-range transport may play key roles in the formation of new particles. ~~Based on our data, we furthermore conclude that NPF may be a frequent and geographically extended phenomenon in the Arctic free troposphere~~In particular, we deduce that one possible mechanism leading to free tropospheric NPF could be associated with warm-air intrusions.

1 Introduction

Aerosol particles are an important component of the global climate system (Forster et al., 2021). They enter the atmosphere either directly by primary emissions or are formed in situ by secondary aerosol particle formation from condensable gases (Seinfeld and Pandis, 2006). The latter process, which generates particles with diameters of initially a few nanometers, is termed new particle formation ~~(NPF) (Kulmala et al., 2004)~~(NPF, Kulmala et al., 2004). Depending on the atmospheric thermodynamic

conditions, subsequent growth can increase particle sizes to values large enough such that the particles may act as cloud condensation nuclei ~~(CCN) (Kulmala and Kerminen, 2008; Kerminen et al., 2018)~~(CCN, Kulmala and Kerminen, 2008; Kerminen et al., 2018). CCN play a crucial role in cloud formation, affecting cloud life time, precipitation formation, and cloud albedo, thereby influencing Earth's radiative energy budget (Andreae and Rosenfeld, 2008; Forster et al., 2021). Globally, about 50 % of CCN are estimated to result from NPF (Gordon et al., 2017; Pierce and Adams, 2009; Merikanto et al., 2009). In the Arctic, with commonly low background aerosol particle number concentrations, NPF is particularly important because of the high susceptibility of Arctic cloud formation (and hence cloud radiative forcing) to changes in aerosol particle and CCN number concentrations under low aerosol levels (Mauritsen et al., 2011; Leaitch et al., 2013; Carslaw et al., 2013; Gordon et al., 2017).

Currently, the Arctic is changing rapidly in response to global climate change (Box et al., 2019; Wendisch et al., 2017, 2023). Among others, these changes manifest in a substantial decrease of the Arctic sea ice cover, especially at the end of the Arctic summer in September (Stroeve et al., 2007). Furthermore, an increase of the near-surface air temperature is observed, which exceeds the global average warming by a factor of two to three (Wendisch et al., 2023). The processes causing the intensified response of the Arctic climate system to global climate change are commonly referred to as Arctic amplification (Serreze and Francis, 2006; Serreze and Barry, 2011; Wendisch et al., 2023). A changing Arctic also entails consequences for the Arctic aerosol population. For example, the decline of the sea ice cover affects both primary emissions from the ocean surface and secondary formation of aerosol particles (Willis et al., 2018; Schmale et al., 2021). Concerning the latter, enhanced emissions of precursor gases from open sea surfaces, leads, or through thinning sea ice have been reported and are expected to proceed as the sea ice fractures and retreats further (Cuevas et al., 2018; Galí et al., 2019). There is ~~direct~~ 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. Furthermore, altered meridional transport may lead to enhanced aerosol particle and/or precursor transport into the Arctic, for instance, via warm-air intrusions (Dada et al., 2022; Wendisch et al., 2023, 2024), which are expected to occur more frequently in the future (Bintanja et al., 2020; Wendisch et al., 2025). 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).

NPF has been observed in several locations Arctic-wide (Willis et al., 2018; Schmale and Baccarini, 2021). Most studies were based on measurements conducted during shipborne campaigns (e.g., Wiedensohler et al., 1996; Allan et al., 2015; Baccarini et al., 2020) or at ground-based stations including Alert, Villum, Zeppelin, Tiksi, and Utqiagvik ~~(e.g., Leaitch et al., 2013; Allan et al., 2015; Croft et al.~~ (e.g., Leaitch et al., 2013; Croft et al., 2016; Asmi et al., 2016; Freud et al., 2017; Beck et al., 2021). These stations provide temporally extended datasets and enable the analysis of annual cycles and long-term trends. Yet, inherently, station- and ship-based measurements can hardly provide precise vertical information. However, data on the vertical distribution of NPF are crucial to identify and study key transport and formation processes, and ultimately, to assess the corresponding effects on the Arctic climate (Lee et al., 2019; Schmale et al., 2021; Price et al., 2023). There are remote sensing instruments such as lidar that can provide vertical information on aerosol particles, but these methods mostly miss the required sensitivity to reliably infer quantitative information on NPF. 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; Kupiszewski et al., 2013; Willis et al., 2016, 2017~~ ~~. These latter airborne studies predominantly suggested that~~ (Garrett et al., 2002; Weber et al., 2003; Engvall et al., 2008; Khosrawi et al., 2

. 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 a
. Hence, the relative importance of the ABL and the free troposphere as sources of newly formed particles remains elusive.
A number of studies reported on the predominant occurrence of NPF close to the surface ~~inside the atmospheric boundary layer (ABL) prevails, and that free tropospheric NPF is less common (Willis et al., 2018)~~(Kupiszewski et al., 2013; Willis et al., 2016, 2017; Buri
, while other measurements detected NPF in the free troposphere as well (Weber et al., 2003; Khosrawi et al., 2010).
In this paper, rather than focusing on the process of NPF itself (i.e., the initial nucleation mechanism), we investigate the occurrence of newly formed particles produced by NPF, which have initial diameters of less than 20 nm (Kulmala et al., 2004). Here, we report on the frequent occurrence of these newly formed particles in the Arctic, found at various altitudes ranging from about 60 m to 3900 m above sea level. The corresponding data were collected during an aircraft campaign carried out in the Fram Strait, surrounding the Svalbard Archipelago, and in the vicinity of Villum Research Station (Station Nord) in Greenland in April 2024. ~~Our data show that newly formed particles can be found throughout the entire lower Arctic troposphere, which indicates that both local processes and long-range transport are important causes for NPF in the Arctic.~~

2 Methods

2.1 Aircraft campaign overview

The airborne campaign "Boundary layer and Aerosol and Cloud Study in the Arctic, based on aircraft and T-Bird Measurements II" (BACSAM II) was conducted from 5 April to 2 May 2024. We carried out eleven research flights (RFs) with the German research aircraft Polar 6 (P6, Wesche et al., 2016), operated by the Alfred Wegener Institute, Helmholtz Centre for Polar and Marine Research (AWI; see supporting information (SI) Fig. S1 for a map of all flight paths). With the operational base in Longyearbyen (Svalbard), target areas included the Fram Strait, the Svalbard Archipelago, and the vicinity of Villum Research Station in north-eastern Greenland. Flights were conducted between 7 and 29 April 2024. This time period was characterized by persistent warm air advection driven by low pressure systems south-west of Svalbard, resulting in above-average surface temperatures in Svalbard. Two exceptions with typical cold conditions and high pressure at the beginning of the campaign from 7 to 11 April and from 15 to 17 April occurred. Note that our sampling is potentially biased towards viable flight conditions, i.e., 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.

2.2 Aerosol measurements and detection of newly formed particles

Aboard the P6 aircraft, we measured aerosol particle number size distributions (PNSDs) with a mobility particle size spectrometer (MPSS, Wiedensohler et al., 2012, 2018). The custom-built MPSS measures the PNSDs in the particle diameter size range between 10 nm and 850 nm based on the electric mobility of aerosol particles with a time resolution of about 300 s. In this paper, we only use data up to 600 nm since on some measurement days arcing inside the differential mobility analyzer

(DMA) led to artifacts in the PNSDs appearing at larger diameters. The system features a Vienna-type DMA (built by the Leibniz-Institute for Tropospheric Research) and a condensation particle counter (CPC, TSI model 3760A, TSI Incorporated, USA). For further details on the MPSS system see Wiedensohler et al. (2012, 2018). The flow in the sampling tubing leading to the MPSS was manually adjusted depending on the true airspeed of the aircraft (typically $\approx 60\ \mathrm{ms}^{-1}$ during legs in the target area and $\approx 90\ \mathrm{ms}^{-1}$ during transit flights) to achieve near-isokinetic sampling. As the onboard-temperature was significantly higher than the ambient temperature, the relative humidity of the sampled air in the aerosol inlet was below 40 %. Consequently, all reported particle diameters refer to dry conditions (Jurányi et al., 2025). All particle number concentration data have been corrected to standard conditions (1013 hPa and 273.15 K). Particle losses inside the sampling line 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).

~~In this paper, we distinguish between NPF and newly formed particles.~~ Since the minimum diameter of aerosol particles that can be detected with our measurement instrumentation is 10 nm, it is not feasible to clearly identify the exact region where NPF occurred, i.e., where the process of the initial nucleation of particles with sizes of about 2 to 3 nm from condensable gases took place (Kulmala et al., 2004; Kerminen et al., 2018). Nevertheless, particle sizes of 10 nm still imply that these particles

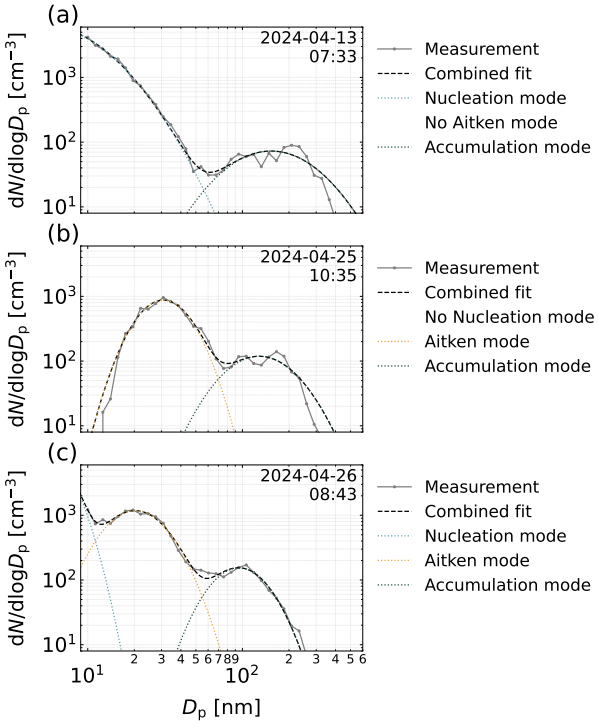


Figure 1. Examples of MPSS-measured PNSDs, showing nucleation and/or Aitken, and accumulation modes. The PNSDs in the panels **(a)** and **(c)** are considered as indicative for newly formed particles, while the measurement displayed in panel **(b)** shows no indication for newly formed particles.

were newly formed by secondary particle formation (i.e., by NPF, Kulmala et al., 2004). We hence refer to these particles as newly formed particles ~~.~~ and, in this paper, instead on the process of NPF, focus on the occurrence of such secondarily formed particles.

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~~. See Sect. S2.1 for further details and examples of fitted PNSDs, indicating either the presence or absence of~~ , i.e., a mode with a maximum below the lower size limit of the MPSS. The detection of newly formed particles ~~(Fig. S2).~~ 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.

2.3 Meteorological measurements

A nose boom was installed at the tip of the aircraft to measure air pressure, temperature, and the three-dimensional wind vector. The nose boom features an Aventech five-hole probe to measure static, dynamic, and differential pressures to derive the wind vector from these data, and an open-wire Pt100 in an unheated Rosemount housing to record air temperature data. These data were measured with a time resolution of 100 Hz. Humidity data are available at 20 Hz from a Vaisala HMT333 in a Rosemount housing installed aboard the aircraft. Surface skin temperature data at 20 Hz are derived from measurements by a KT-19 infrared pyrometer (Heitronics KT-19.85 II). Position and altitude were measured by combining Global Positioning System (GPS) and Inertial Navigation System (INS) data, which were recorded at 100 Hz. See Hartmann et al. (2018) for further details on the instrumentation and data processing.

2.4 Backwards trajectory analysis

Backward trajectories were calculated from ERA5 wind fields (Hersbach et al., 2020) utilizing the Lagrangian analysis tool LAGRANTO (Sprenger and Wernli, 2015). The trajectories were initiated every minute along the flight track of the P6 at the corresponding flight altitude. The trajectories were calculated for 120 hours backwards in one minute time steps and several meteorological parameters from ERA5 were traced along the trajectory. Based on these parameters, we carry out an analysis of air mass history in Sect. 3. Among others, we determine the last contact of these air masses with clouds based on the specific cloud liquid water content ($CLWC$) and the specific cloud ice water content ($CIWC$). Here, we consider an air mass to have been in contact with a cloud if ~~$CIWC > 0.01\,\mathrm{g\,kg^{-1}}$ or $CLWC > 0.1\,\mathrm{g\,kg^{-1}}$.~~ the total water content ($CWC = CLWC + CIWC$) exceeds the threshold of $CWC > 0.01\,\mathrm{g\,kg^{-1}}$ (McFarquhar and Cober, 2004; Moser et al., 2023, see Sect. S3).

3 Results and discussion

3.1 ~~Frequent occurrence~~ Geographical and vertical distribution of newly formed particles

Throughout the entire BACSAM II campaign, we frequently observed newly formed particles over large areas of the Arctic region studied (Fig. 2). Newly formed particles were detected during all eleven research flights, with the first observation of such particles on 7 April 2024, and the last on 29 April 2024. We encountered newly formed particles at various locations (approximately between 76°N to 85°N and 15°W to 26°E) and altitudes ranging from about 60 m to 3900 m above sea level (Fig. 2). The time periods during which we identified newly formed particles amount to an overall duration of about 10 hours and 30 minutes, which corresponds to approximately ~~24~~29 % of the total ~~flight time~~ measurement time (at constant altitude) of all research flights and a distance flown of about ~~2800 km.~~ 2770 km.

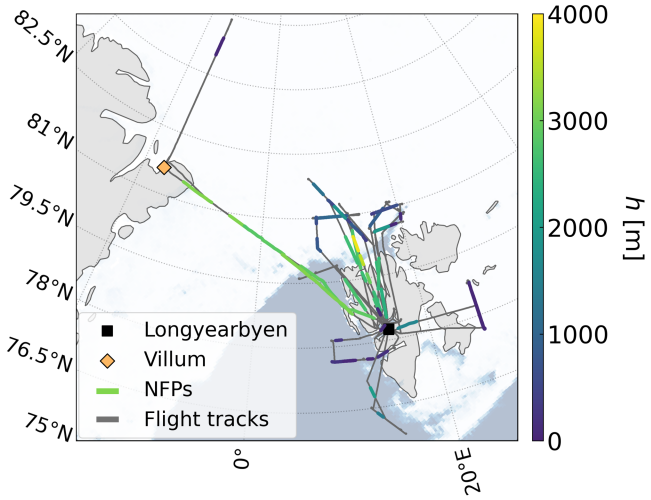


Figure 2. Geographical location and altitude at which newly formed particles (NFPs) were observed during the BACSAM II campaign. The flight tracks are shown in gray and the locations where newly formed particles were identified are color-coded by the corresponding flight altitude. The sea ice concentration is shown exemplary for 15 April 2024, during which we encountered newly formed particles at low altitudes over the ice-covered sea east of Svalbard (see Sect. 3.3). The corresponding sea ice data were retrieved from www.seaice.uni-bremen.de (Spreen et al., 2008).

~~We categorize the observations according to three~~ 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. 3). 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), ~~and~~ newly formed particles in the vicinity of clouds (Sect. 3.4)~~.~~ ~~The reader is referred to Fig. S3 for the~~ , 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. 3a depicts the median size distributions of the PNSDs indicating the presence of newly formed particles, for each of the ~~three environments. Furthermore, we report 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~~in Sect~~. 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. ~~S3 and Fig. S4.~~

~~Newly formed particles~~ About 45 % of all PNSDs featuring newly formed particles were detected in the free troposphere~~account for the majority of our observations with about 45~~, corresponding to approximately 4 hours and 40 minutes of measurements (Fig. 3b, c) and distances flown of $\approx 1460\,\mathrm{km}$. About 16 ~~% of all cases (i.e., 45 % of all PNSDs~~ ($\approx$ 1 hour and 40 minutes, $\approx 350\,\mathrm{km}$) of all cases featuring newly formed particles ~~), compared to about 16~~were found in the ABL over sea ice, as well as 6 % ($\approx$ 40 minutes, $\approx 130\,\mathrm{km}$) in the ABL over ~~sea ice, and approximately~~ ocean with sea ice floes. Approximately 25 % ($\approx$ 2 hours and 40 minutes, $\approx 630\,\mathrm{km}$) of all cases were observed in the vicinity of clouds (Fig. 3~~a). About 14~~b, c). For approximately 8 % ~~of all observations cannot~~ ($\approx$ 50 minutes, $\approx 200\,\mathrm{km}$) 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 ~~(Sect. S2.2). Newly~~ 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).

As mentioned earlier, newly formed particles were ~~frequently~~ detected in a wide range of altitudes between approximately 60 m and 3900 m (Fig. 3~~b, c~~b-d). Here, 60 m and 3900 m correspond to the minimum and maximum flight altitudes at which measurements were carried out ~~.~~ (Fig. 3b). Consequently, we cannot rule out that newly formed particles could be present at altitudes even higher than 3900 m ~~. In~~ in the target regions during our campaign. Fig. 3~~c, we show the relative~~ d presents an overview of the frequency of occurrence of newly formed particles ~~per altitude~~during our campaign. Here, we show the relative frequency of occurrence per altitude bin, defined as the duration of newly formed particle-observations per altitude, normalized by the total time of all PNSD measurements within this altitude bin. The frequency values show a rather ~~uniform vertical distributionfrom the surface up to about 2000 m, with~~ variable vertical distribution. For example, in the lowest 300 m, roughly 25 % of all measured PNSDs ~~featuring~~ featured newly formed particles. At altitudes between about 2500 m and 3200 m, ~~where measurements were overwhelmingly carried out in the cloud-free free troposphere (Fig. 3b), newly formed particles were detected more~~

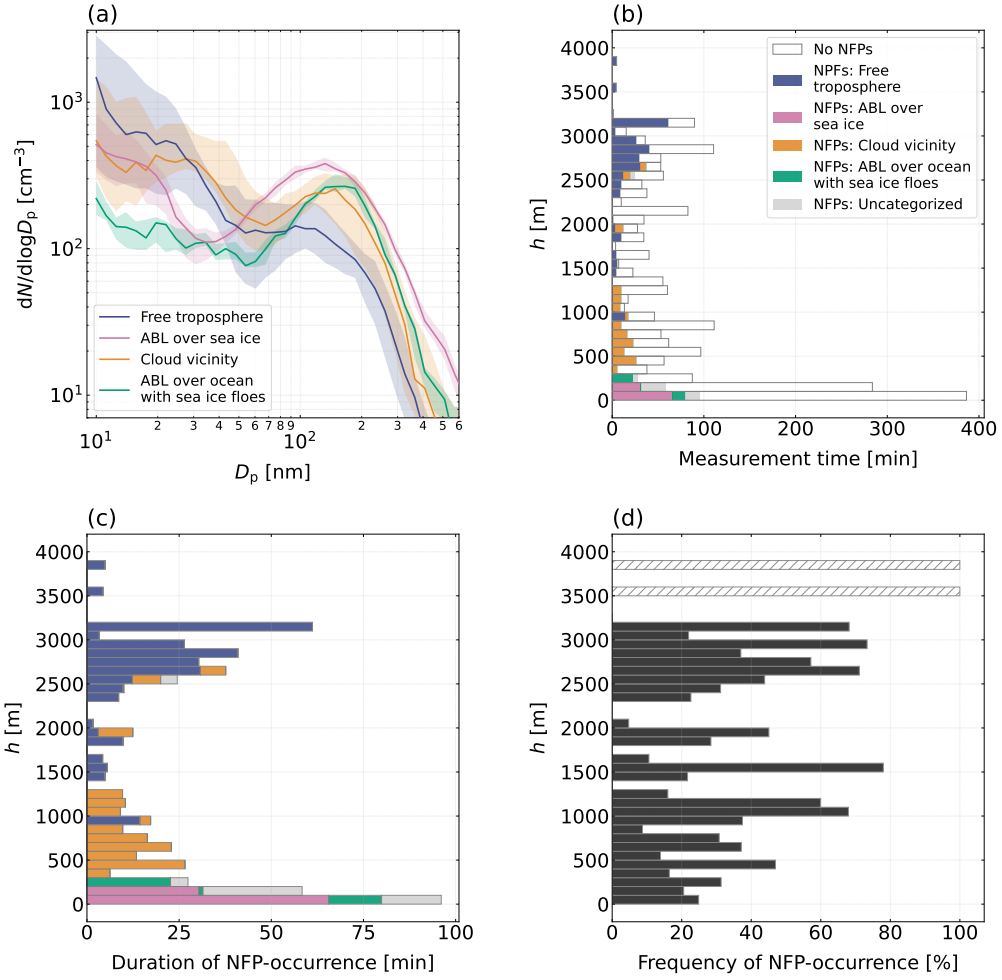


Figure 3. **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and inside 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. 3a).

frequently, with up to 67 up to almost 75 % of PNSDs measured at these altitudes indicating indicate the presence of newly formed particles (Fig. 3c). To additionally provide a general overview of particle number concentrations prevailing throughout the campaign, Fig. 3d presents the median vertical distribution of particle number concentrations in the total measurement size range from 10 nm to 600 nm and in the nucleation mode size range from 10 nm to 20 nm, calculated from the data available from all flights. The values span roughly two orders of magnitude, with nucleation mode particles, e.g., contributing between 50 % to 75 % of all measured particles at altitudes of about 1000 m and 3000 m.d). Note that during the campaign, sampling times and consequently number of PNSDs measured, as well as distances flown at certain altitudes, were not evenly distributed over the investigated altitudes, geographical locations, and synoptic conditions . (Fig. 3b). For example, while measurements in the lowest 100 m were carried out for a total duration of approximately 390 almost 400 min (over a distance of about 1480 km), measurements at 3500-3600 m and 3800-3900 m were solely performed during one research flight, and at each altitude range for merely 5 to 10 min (27 to 54 km, to feature one full MPSS scan; hatched bars in Fig. 3c). d). The values of duration and relative frequency of newly formed particle-occurrence reported in this section are thus not intended to constitute representative values for the lower Arctic troposphere, but rather . They rather provide a rough overview and represent statistics of the observations made during the BACSAM II campaign. **(a)** Duration of newly formed particle-occurrence (NFP-occurrence) for each of the categories, which are introduced in detail in the main text (left y-axis). Ratio of NFP-observations per category to all NFP-observations combined (right y-axis). **(b)** Duration of NFP-occurrence per measurement altitude, grouped into height bins with a width of 100 m. The data are shown as stacked bars. **(c)** Relative frequency of NFP-occurrence per measurement altitude, grouped into height bins with a width of 100 m. The hatched bars at altitudes higher than 3500 m indicate that here only one full PNSD measurement was conducted within each altitude bin. **(d)** Median values (solid lines) and interquartile ranges (shaded area) of integral particle number concentrations in the total measured size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 600\ \mathrm{nm}$) and in the nucleation mode size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 20\ \mathrm{nm}$), respectively. Despite these limitations, we additionally present statistics concerning the total measurement times spent in the free troposphere, in the ABL over sea ice and over ocean with sea ice floes, and in cloud vicinity throughout the campaign in Sect. S3, based on ERA5 reanalysis data. Further, relative frequencies of occurrence of newly formed particles for these environments are provided. The values are associated with rather large uncertainties and can therefore only provide a very rough estimate (see Sect. S3 for details).

Nevertheless, our measurements clearly demonstrate that newly formed particles can frequently be were frequently found throughout the lower Arctic troposphere and , as these particles originate from secondary particle formation, suggest that NPF in the Arctic occurs occurred across large geographical areas and over a wide range of altitudes . In this context, it is worth noting that no indications for the presence of newly formed particles were found within or near the cloud-free ABL over open ocean (Sect. S2.2). during our campaign in April 2024.

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 three 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 the these three environ-

ments. 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.

3.2 Newly formed particles in the free troposphere

Here, as an illustrative example, we present results concerning newly formed particles in the free troposphere which were encountered on 13 April 2024 during RF04 while transiting from Longyearbyen (Svalbard) to Villum Research Station (northern Greenland) at an altitude of 2890 m (Fig. 4a).

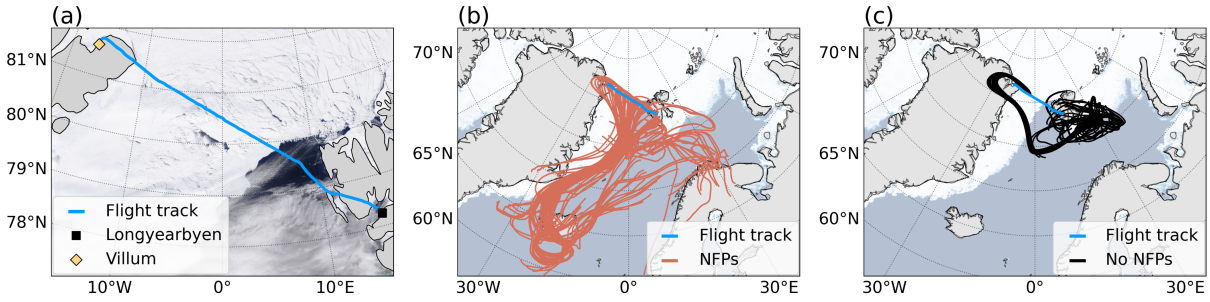


Figure 4. (a) Flight track of RF04 on 13 April 2024. The Moderate-resolution Imaging Spectroradiometer (MODIS)-TERRA satellite image was retrieved from https://wvs.earthdata.nasa.gov. **(b)** Five-day backward trajectories ~~along the flight track. Backward trajectories~~ associated with newly formed particles (NFPs) ~~are depicted~~ along the section of the flight track shown in ~~red and~~ Fig. 5. **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles~~in black~~. The sea ice concentration data shown in the subplots (b) and (c) were retrieved from www.seaice.uni-bremen.de (Spreen et al., 2008).

In the two days ahead of the flight, a low pressure system located over the east coast of Greenland advected warm and moist air over the sea ice edge in the Fram Strait (~~red trajectories in~~ Fig. 4b). Within this warm-air intrusion, North Atlantic ABL air was lifted up to the free troposphere. The uplifting over the sea ice edge was accompanied with the formation of precipitating clouds, which dissipated before sampling of the air mass. Consequently, the sampled free tropospheric air was cloud-free. The cumulus clouds visible in the satellite image in Fig. 4a capped the marine ABL, which was located more than 2 km beneath the flight track. Therefore, we expect no influence of these low-level clouds on our measurements in the free troposphere. In addition to the southerly air masses from the Atlantic, air masses originating at higher latitudes close to Svalbard were probed during RF04 (~~black trajectories in~~ Fig. 4~~b). **(a)** P6 altitude during RF04 on 13 April 2024. **(b)** MPSS-measured PNSD. **(c)** Integral particle number concentrations in the total measured size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 600\ \mathrm{nm}$, solid line) and in the nucleation mode size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 20\ \mathrm{nm}$, dashed line), respectively.~~ c).

During the time period from about 6:23 to 7:40 UTC, flying at high altitudes ($\approx$ 2890 m), and covering a distance of about

409 km, we encountered elevated number concentrations in the lower MPSS size bins (Fig. 5). The measured PNSDs featured a sharp decline in the size range from 10 nm to about 60 nm, i.e., we observed the "upper" end of a pronounced nucleation mode (see Fig. S8 for the related median PNSD). Detected particle number concentrations in the size range from 10 nm to 20 nm reached on average values of about ~~$N_{10-20} \approx 714\ \mathrm{cm}^{-3}$~~ $N_{10-20} \approx 778\ \mathrm{cm}^{-3}$, which constituted about ~~66~~68 % of the total particle number concentration measured in the range from 10 nm to 600 nm (Fig. 5c). ~~The measured PNSDs featured a sharp decline in the size range from 10 nm to about 60 nm, i.e., we observed the "upper" end of a pronounced nucleation mode (see Fig. S5 for the related median PNSD).~~ In other words, we detected newly formed particles in the Arctic free troposphere across a large geographical area.

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 (~~$N_{10-20} \approx 25\ \mathrm{cm}^{-3}$, 16~~$N_{10-20} \approx 27\ \mathrm{cm}^{-3}$, 17 % of the total MPSS-measured number concentration, Fig. 5c), i.e., no ~~or only a small number of~~ newly formed particles were present. Notably, the newly formed particles observed during the first part of the transit flight were associated with air masses originating from lower latitudes which were transported into the Arctic within a warm-air intrusion (~~red trajectories in~~ Fig. 4b), while ~~for~~ in the northerly air masses no newly formed particles were detected (~~black trajectories in~~ Fig. 4~~b~~c).

~~The presence of newly formed particles in the Arctic free troposphere distinguishes our observations from previous airborne studies utilizing aircraft and helicopter measurements, which pointed towards NPF within the ABL as the dominant source of newly formed particles in the Arctic (Engvall et al., 2008; Kupiszewski et al., 2013; Burkart et al., 2017; Willis et al., 2016, 2017) . Together with a number of ground-based studies highlighting the importance of NPF within the ABL (e.g., Ström et al., 2009; Leaitch et al , these results led researchers to assume that NPF in the Arctic primarily occurs close to the surface (e.g., Willis et al., 2018; Dall'Osto et al.,~~

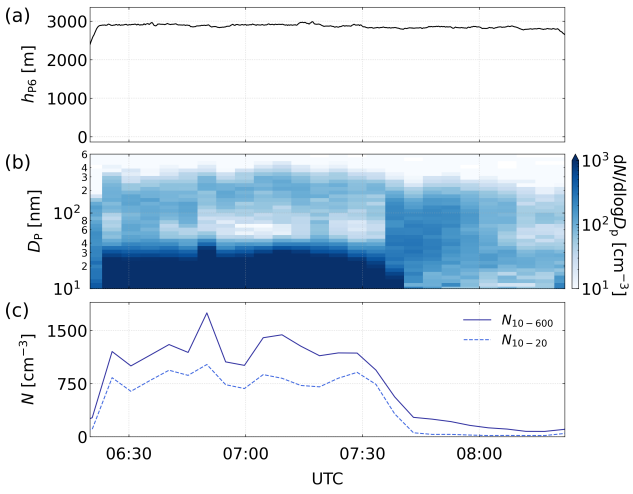


Figure 5. **(a)** P6 altitude during RF04 on 13 April 2024. **(b)** MPSS-measured PNSD. **(c)** Integral particle number concentrations in the total measured size range (10 nm $\leq D_p \leq$ 600 nm, solid line) and in the nucleation mode size range (10 nm $\leq D_p \leq$ 20 nm, dashed line), respectively.

~~. However, high numbers of newly formed particles in the Arctic free troposphere have occasionally been reported before (Wiedensohler et al., 1996; Weber et al., 2003). 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. During the other 37 flights of their campaign, no indications for NPF occurring at high altitudes were found. 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. Our data support these latter observational results and indicate that NPF might occur frequently in the Arctic free troposphere over large geographical areas.~~

Backward trajectory analysis of all air masses featuring newly formed particles in the free troposphere

To further elucidate free tropospheric newly formed particles, which we observed on eight different days, considering all research flights performed during BACSAM II, and utilizing backward trajectories, a systematic analysis of free tropospheric air mass history was carried out. We investigated both free tropospheric air masses that show clear signs of newly formed particles, and free tropospheric air masses that exhibit no indications of newly formed particles. Every minute of each flight, we initiated a five-day backward trajectory at the pressure altitude of the aircraft. Hence, the air sampled during one PNSD measurement, which lasts for approximately 5 minutes and indicates either the presence or absence of newly formed particles, is represented by five backward trajectories. The results of the analysis are presented in Fig. 6, which depicts median values of meteorological and geographical parameters along the trajectories of all flight sections carried out in the free troposphere throughout the campaign.

Predominantly, air masses featuring newly formed particles in the free troposphere originated further south compared to air masses with no indications for free tropospheric newly formed particles, and gradually moved poleward at high altitudes during the five days prior to arrival (Fig. 6a-c). Initially, these air masses connected to newly formed particle were warmer and contained more moisture (Fig. 6d, f). Higher values of northward integrated water vapor transport (IVT_{north}) show that in the whole column more water vapor was transported poleward for cases when newly formed particles were observed in the Arctic free troposphere (Fig. 6g). 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~~. The latter might be caused by the formation of (precipitating) clouds, as already mentioned in the case study above~~, 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. ~~Generally, about~~ 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

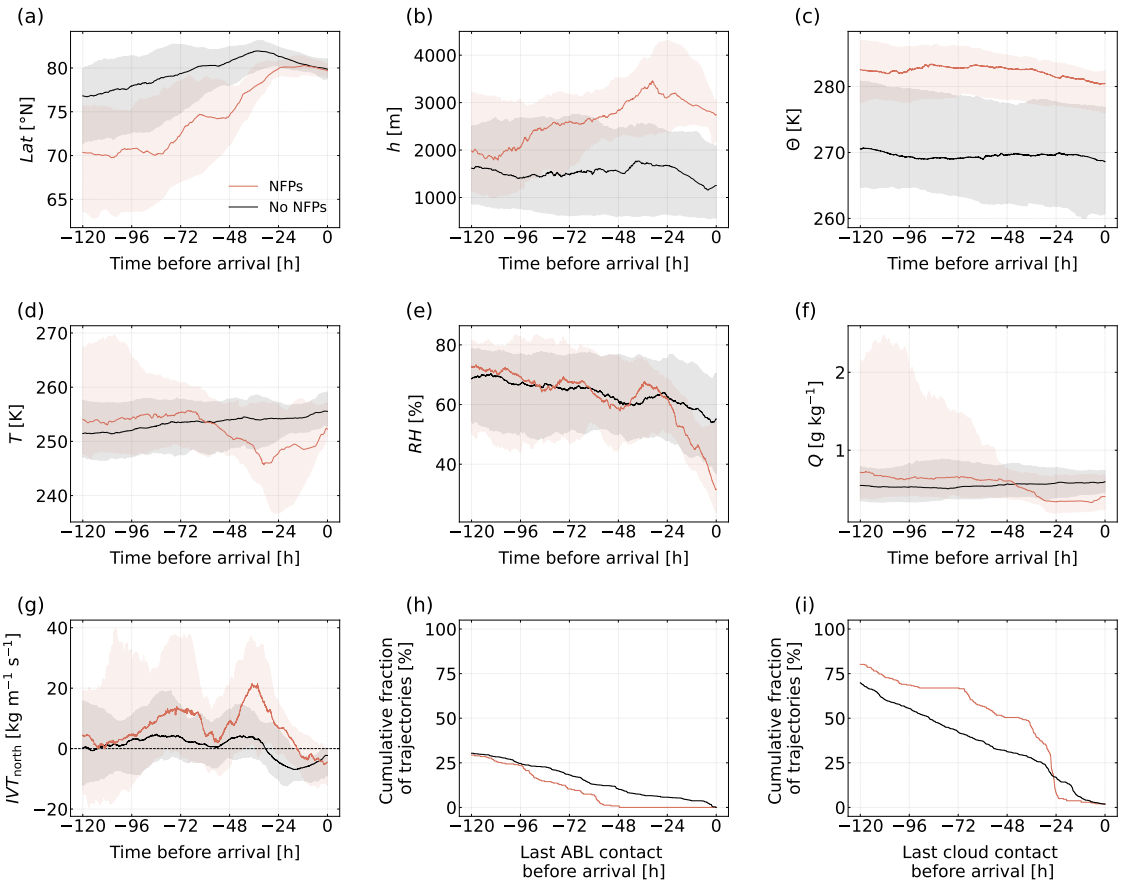


Figure 6. Comparison of geographical and meteorological parameters along the backward trajectories of flight sections in the free troposphere between trajectories connected to the presence (red) and absence (black) of newly formed particles (NFPs). Shown, as a function of time prior to arrival at the measurement location, are median values (solid lines) and interquartile ranges (shaded areas) of **(a)** latitude Lat, **(b)** altitude h, **(c)** potential temperature Θ, **(d)** absolute temperature T, **(e)** relative humidity RH, **(f)** specific humidity Q, and **(g)** northward integrated water vapor transport IVT_{north}. The last two subplots show the cumulative fraction of the last contact of the trajectories with the **(h)** ABL and **(i)** clouds. Note that if the cumulative fraction does not increase to 100 % at $t = -120\,\mathrm{h}$, this indicates that a fraction of the trajectories was not in contact with the ABL/clouds during the last five days (the maximum time span the calculated trajectories cover).

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 ~~influenced by~~ in contact with clouds on their way to the measurement location ~~(Fig. 6i). In contrast, only 28~~during the previous five days, as well as about 70 % of the backward trajectories featuring no newly formed particles ~~were in contact with a cloud during the last five days before arrival~~ (Fig. 6i). ~~Higher values of northward integrated water vapor transport (IVT_{north}), most notably~~ However, no clouds were present during the last ~~12 to 48 hours , show that in the whole column more water vapor was transported poleward for cases when~~ 24 hours before the measurement

for the air masses featuring newly formed particles~~were observed in the Arctic free troposphere~~ . 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. 6~~g)~~ f) already mentioned above indicates that the removal of clouds, at latest 24 hours prior to arrival, could (partly) be caused by precipitation. None of the backward trajectories featuring newly formed particles resided in the ABL for the last 12 hours prior to arrival at the track of the P6, and less than 2 % of these trajectories passed through the ABL in the last 54 hours. About ~~71~~70 % of the backward trajectories had no contact with the ABL for the last five days (Fig. 6h).

These findings suggest that the cases of newly formed particles in the Arctic free troposphere ~~are~~ observed during our campaign are predominantly connected to poleward transport of warm and moist air, as occurring, e.g., during warm-air intrusions (Wendisch et al., 2024). Furthermore, because the trajectories overwhelmingly remained in the free troposphere during the transport to the measurement region, the results indicate that the precursor gases involved in the formation of these particles originated from long-range transport from lower latitudes, with NPF very likely occurring in the Arctic free troposphere. Such a transport pathway for precursor gases resulting in free tropospheric NPF has already been suggested in a modeling study by Price et al. (2023). ~~Furthermore, the~~

The above findings imply a potential role for clouds in the transport and possibly 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).

~~This leads~~ 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 helicopter (Kupiszewski et al., 2013) and aircraft measurements (Burkart et al., 2017; Willis et al., 2016, 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); 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.

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.

3.3 Newly formed particles inside the atmospheric boundary layer over sea ice

Inside the ABL, newly formed particles were detected, e.g., on 15 April 2024 during RF05 while flying east of Svalbard at low altitudes (approximately 70 m and 155 m) over sea ice, where cracks as well as open and partly refrozen leads were present (~~see Fig. S6~~Fig. S9). The associated flight track is shown in Fig. 7a, with Fig. 7b, c depicting the backward trajectories for the air masses encountered along the flight track.

The area investigated was subject to the anti-cyclonic flow around a high pressure system north of Svalbard, causing cold air advection from the central Arctic to the target region. This meteorological situation resulted in air masses that exclusively originated in the high Arctic and traveled ~~towards~~ toward the P6 flight track from higher latitudes over sea ice-covered regions (Fig. 7b, c). All the way between waypoint 1 (WP1) and WP3, over a distance of about 115 km, clear sky conditions prevailed (Fig. 7a).

The presence of newly formed particles ~~was indicated by~~ is indicated by the occurrence of a distinct nucleation mode, i.e., ele-

vated aerosol particle number concentrations in the lower size bins of the PNSDs, ~~featuring a distinct nucleation mode,~~ which we observed during three consecutive legs at altitudes of ≈ 70 m and ≈ 155 m between WP1 and WP3 (Fig. 8). Mean number concentrations in the nucleation mode size range reached ~~$N_{10-20} \approx 191\ \mathrm{cm}^{-3}$~~ $N_{10-20} \approx 206\ \mathrm{cm}^{-3}$ (time period 10:03 to 10:39 UTC), ~~$N_{10-20} \approx 170\ \mathrm{cm}^{-3}$~~ $N_{10-20} \approx 185\ \mathrm{cm}^{-3}$ (11:08 to 11:22 UTC), and ~~$N_{10-20} \approx 114\ \mathrm{cm}^{-3}$~~ $N_{10-20} \approx 125\ \mathrm{cm}^{-3}$ (12:17 to 12:40 UTC), accounting for about ~~39~~40 %, ~~37~~38 %, and ~~30~~31 % of the total MPSS-measured aerosol particle number concentrations. In contrast, during the leg at ≈ 850 m altitude (11:30 to 12:00 UTC) between WP1 and WP3, as well as during the transit to and from the target region at ≈ 1360 m altitude, the PNSDs featured no nucleation mode and, accordingly, low number concentrations in the ~~nucleation mode~~ sub-20 nm size range (Fig. 8, $N_{10-20} \approx 3\ \mathrm{cm}^{-3}$, 1 % of the total MPSS-measured particle number concentration at 850 m). From the vertical profiles of potential temperature obtained during the ascents and descents at WP1, WP2, and WP3 (Fig. 8a), we determine an ABL height of about ~~170~~ 190 m in the region of interest (see Fig. ~~S7~~S10). Thus, the three legs at 70 m and 155 m, during which newly formed particles were detected, were carried out inside the ABL, while the leg at higher altitude (≈ 850 m), as well as the transits to and from the region of interest, took place in the free troposphere.

Integral aerosol particle number concentrations in the total measured size range (10 nm to 600 nm) and in the size range from 10 nm to 20 nm decreased from WP1 to WP3, i.e., from south to north (Fig. 7a, Fig. 8~~c).~~). The spatial variability of the number concentration of nucleation mode particles ~~suggests geographically heterogeneous processes and~~ points toward newly formed particles as the result of regional formation rather than long-range transport~~, as the latter would likely lead to rather constant concentrations of newly formed particlesalong the flight track.~~ . Nucleation mode particles have short atmospheric residence times due to their high diffusion, coagulation with larger particles, and condensational growth out of the nucleation mode size (e.g., Pruppacher and Klett, 2010; Wallace and Hobbs, 2006). These effects tend to smooth out

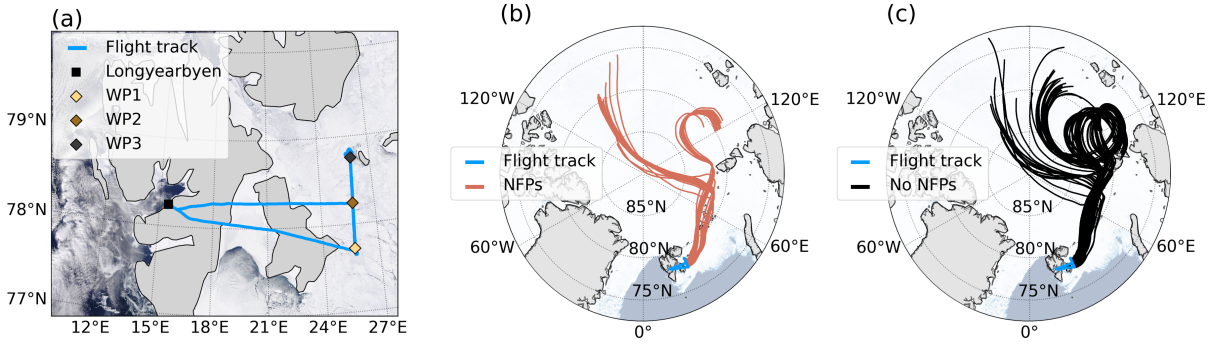


Figure 7. (a) Flight track of RF05 on 15 April 2024. The MODIS-TERRA satellite image was retrieved from https://wvs.earthdata.nasa.gov. **(b)** Five-day backward trajectories associated with newly formed particles (NFPs) along the section of the flight track shown in Fig. 8. ~~Backward trajectories associated with newly formed particles (NFPs) are depicted in red and~~ **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles~~in black~~. The sea ice concentration data shown in the subplots (b) and (c) were retrieved from www.seaice.uni-bremen.de (Spreen et al., 2008).

concentration gradients of such small particles, thereby making a contribution from long-range transport to the observed events less likely (Kupiszewski et al., 2013). Interestingly, the particle number concentrations in the nucleation mode size range from 10 nm to 20 nm recorded during the legs inside the ABL between WP1 and WP3 (i.e., in the same area) decreased not only spatially, but also over time (Fig. 8c). In addition to the integral values shown above, Fig. ~~S8~~ S11 depicts median PNSDs for each of the three legs close to the surface individually, which likewise shows that number concentrations of particles in the nucleation mode reduce over time. This temporal variability points toward a regional source as well, since it suggests that we observed an NPF event that becomes weaker throughout the day. However, we are not able to unambiguously resolve if the precursors involved in NPF originated directly from the sea ice-covered measurement region, with various cracks and leads being present (Fig. ~~S6~~S9), or if the precursors were transported to the target area from the higher Arctic. But, in our view, it appears feasible to conclude that the spatial and temporal variability of the newly formed particles encountered here (i.e., the decrease of sub-20 nm particles from WP1 to WP3 and throughout the day) and the similar origin of air masses featuring newly formed particles (Fig. 7b) and air masses without newly formed particles (Fig. 7~~b~~c) rather indicate a regional source.

~~In the Arctic, precursor gases reported to contribute to NPF and particle growth include dimethylsulfide (DMS, which oxidizes to methansulfonic acid (MSA) and sulfuric acid (SA)), ammonia, organics, and iodic acid (Willis et al., 2018; Schmale and Baccarini, 2021) . The latter, in particular, has been linked to NPF in Arctic sea ice-covered regions (Allan et al., 2015; Sipilä et al., 2016; Baccarini et al., 20 . It has been shown for both the Arctic and Antarctica that iodine can be emitted from the sea ice. Potential sources include~~

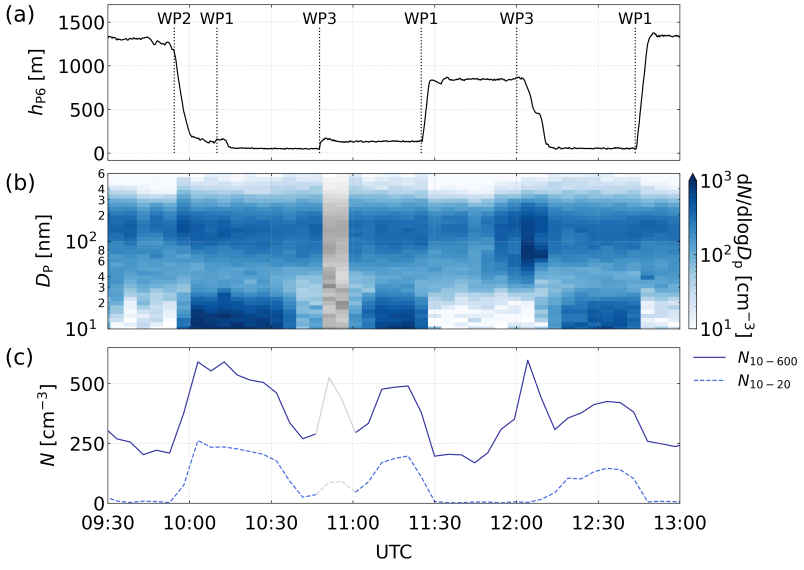


Figure 8. **(a)** P6 altitude during RF05 on 15 April 2024. **(b)** MPSS-measured PNSD. **(c)** Integral particle number concentrations in the total measured size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 600\ \mathrm{nm}$, solid line) and in the nucleation mode size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 20\ \mathrm{nm}$, dashed line), respectively. The data measured from 10:51 to 11:01 UTC are grayed out in panels (b) and (c) since erroneous measurements due to possible sampling of aircraft exhaust cannot be ruled out.

~~marine microalgae (diatoms) growing below the sea ice (Hill and Manley, 2009; Atkinson et al., 2012), where the iodine can be~~
~~emitted through cracks and leads and has been connected to thinning Arctic sea ice (Cuevas et al., 2018). The sea ice itself or the~~
~~snowpack can also release iodine via abiotic, photo-chemical processes (Gálvez et al., 2016; Kim et al., 2016; Raso et al., 2017)~~
~~. Yet, since we cannot resolve the particle composition of nucleation mode particles nor conducted gas-phase measurements,~~
~~at this point, we cannot ultimately determine the precursor gases resulting in the newly formed particles observed in this case~~
~~study. Nevertheless, these previous results suggest the sea ice-covered target area as a plausible source region of precursor~~
~~gases and newly formed particles.~~

Backward trajectory analysis of all air masses featuring newly formed particles in the atmospheric boundary layer over sea ice

To further investigate the occurrence of newly formed particles inside the ABL over sea ice, an analysis of air mass history was carried out for the respective observations made during our campaign. Newly formed particles inside the ABL over sea ice were found twice, i.e., on 15 April (case study) and on 13 April for approximately 15 min north of Villum Research Station (see Fig. 2). Fig. 9 presents the results of the backward trajectory analysis carried out for both ~~days~~events.

Both days were characterized by cold air masses from northern latitudes which traveled over sea ice-covered regions (median sea ice concentration values between ≈ ~~93~~95 % and 100 %, Fig. 9a, b, e). The air masses featured low specific humidities, but, due to cold temperatures, high relative humidities (w.r.t. water, Fig. 9e-g). Furthermore, the data suggest that all trajectories resided inside the ABL during the last 24 hours (Fig. 9i) before arriving at the P6, providing the possibility for precursor uptake. ~~A difference between the two air mass histories is the time since the~~ For about 60 % of the trajectories, the last cloud contact ~~before arrival~~ occurred approximately 24 h before arrival in the target region (Fig. 9j). ~~The air masses encountered on 13 April (RF04) had no cloud contact for at least four days, while during RF05 on~~ Roughly 15 ~~April, last cloud contact of more than 50~~ % of the trajectories ~~occurred about 24 h before arrival in the target region.~~ were not influenced by clouds on their way to the measurement location during the last five days, including 80 % of all trajectories on 13 April. For both days, the trajectories descended during the last five days~~. This decrease in altitude is more pronounced for the event on 15 April (Fig. 9c)~~, with the lowest altitude along the trajectories reached just before arrival at the aircraft ~~.~~ (Fig. 9c). The descent of the backward trajectories again indicates rather regional precursor sources as the gases are very likely emitted from the surface.

~~From these~~ NPF in the Arctic ABL in sea ice-covered regions has been observed e.g. by Allan et al. (2015); Sipilä et al. (2016); Baccarini et . In these studies, concurrent chemical measurements suggested a link between iodine species and NPF events. Furthermore, it has been shown for both the Arctic and Antarctica that iodine can be emitted from the sea ice. Potential sources include marine microalgae (diatoms) growing below the sea ice (Hill and Manley, 2009; Atkinson et al., 2012), where the iodine can be emitted through cracks and leads and has been connected to thinning Arctic sea ice (Cuevas et al., 2018). The sea ice itself or the snowpack can also release iodine via abiotic, photo-chemical processes (Gálvez et al., 2016; Kim et al., 2016; Raso et al., 2017) . Yet, since we cannot resolve the particle composition of nucleation mode particles nor conducted gas-phase measurements, at this point, we cannot ultimately determine the precursor gases which could have participated in the formation of the newly formed particles observed in this case study. Nevertheless, these previous results suggest the sea ice-covered target area as a plausible source region of precursor gases and newly formed particles.

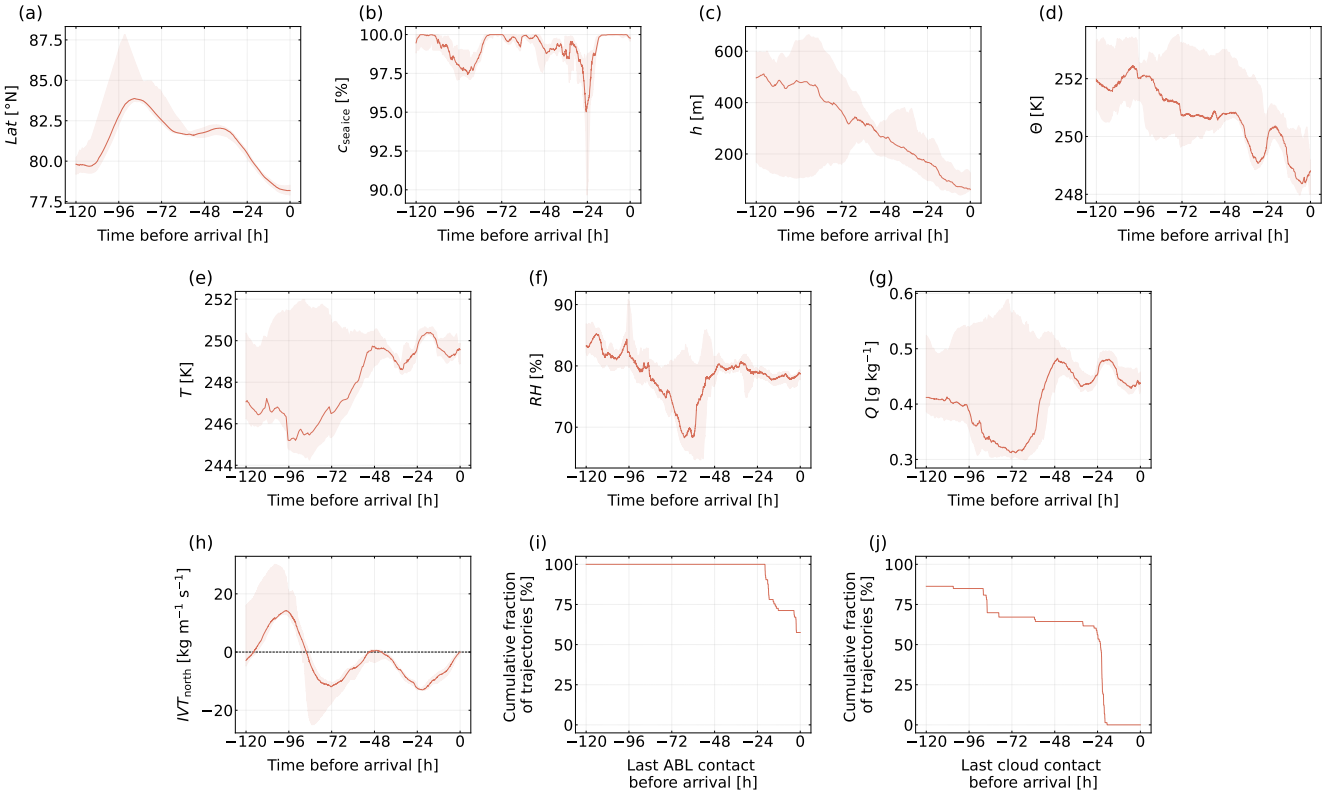


Figure 9. Comparison of geographical and meteorological parameters along the backward trajectories of flight sections linked to newly formed particles inside the ABL over sea ice for the events on 13 and 15 April. Shown, as a function of time prior to arrival, are median values (solid lines) and interquartile ranges (shaded areas) of **(a)** latitude Lat, **(b)** sea ice concentration c_{seaice}, **(c)** altitude h, **(d)** potential temperature Θ, **(e)** absolute temperature T, **(f)** relative humidity RH, **(g)** specific humidity Q, and **(h)** northward integrated water vapor transport IVT_{north}. The last two subplots show the cumulative fraction of the last contact of the trajectories with the **(i)** ABL and **(j)** clouds. Note that if the cumulative fraction does not increase to 100 % at $t = -120$ h, this indicates that a fraction of the trajectories was not in contact with the ABL/clouds during the last five days (the maximum time span the calculated trajectories cover).

In summary, from our observations, we hypothesize that the observed presence of newly formed particles in the ABL over sea ice is connected to precursor gases ~~(conceivably iodine components)~~ (conceivably iodine components, as reported by Allan et al., 2015; Sip which originate from natural, most likely regional, but possibly also higher-latitude Arctic sources within the sea ice, from leads, or snow.

3.4 Newly formed particles in the vicinity of clouds

Newly formed particles in the vicinity of clouds were, e.g., encountered on 24 April 2024 during RF08, while flying north of Svalbard over sea ice (open leads and cracks were present, see Fig. ~~S9~~S12) with an initially slightly broken and then closed

stratocumulus cloud deck (Fig. 10a). Measurements were carried out below cloud base and at cloud top.

On 24 April, a high pressure system was centered directly over Svalbard, resulting in westerly flow in the target region. The air mass probed at altitudes between approximately 600 m and 1200 m originated either in the North Atlantic close to Greenland, near Iceland, or northern Scandinavia (~~red trajectories in~~ Fig. 10b), and was advected from the North Atlantic marine ABL and lifted above the sea ice, where this layer was decoupled from the surface (Sect. ~~S6~~S8.2, Fig. ~~S10~~S13). At the top of that elevated mixed layer, stratocumulus clouds were present (Fig. 10a), with a cloud base at about 800 m altitude, and cloud top at about 1150 m. The air mass that was probed at about 2100 m in the free troposphere originated in northern Canada and passed over Greenland on its way to the P6 position (~~black trajectories in~~ Fig. 10~~b~~c).

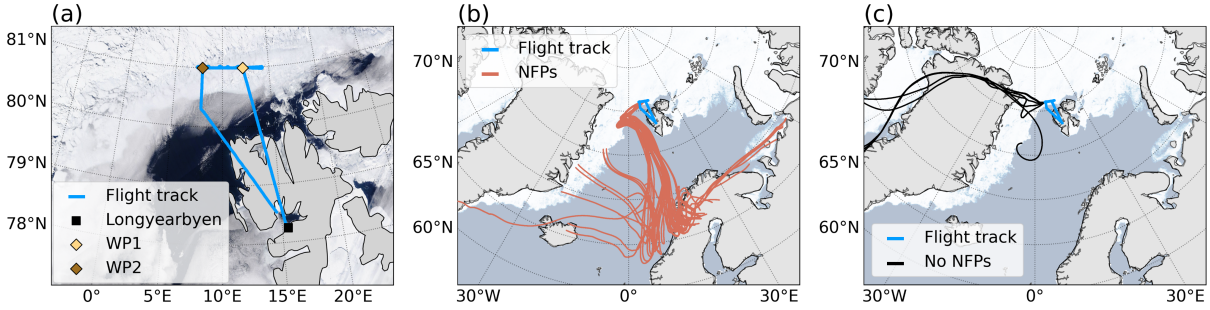


Figure 10. (a) Flight track of RF08 on 24 April 2024. The MODIS-TERRA satellite image was retrieved from https://wvs.earthdata.nasa.gov. **(b)** Five-day backward trajectories associated with newly formed particles (NFPs) along the section of the flight track shown in Fig. 11. ~~Backward trajectories associated with newly formed particles (NFPs) are depicted in red and~~ **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles~~in black~~. The sea ice concentration data shown in the subplots (b) and (c) were retrieved from www.seaice.uni-bremen.de (Spreen et al., 2008).

Here, we specifically discuss the data collected between WP1 and WP2 over a distance of 75 km in the time period between 10:28 and 11:35 UTC. This time period includes a descent from approximately 2100 m down to below cloud level (10:29 to 10:35 UTC), a leg at cloud base (10:35 to 10:52 UTC), an ascent through cloud with subsequent drying away of the ice collected inside cloud, and finally a leg at cloud top (11:07 to 11:35 UTC). With respect to the aerosol particle measurements, the time period 10:52 to 11:07 UTC, i.e., the ascent through cloud and the subsequent drying away of ice, is excluded from further analysis, as erroneous measurements due to icing of the aerosol inlet and/or droplet shattering (Weber et al., 1998) cannot be ruled out. The MPSS data from this period are grayed out in Fig. 11b, c.

The PNSDs at cloud top and at cloud base featured a pronounced nucleation mode and correspondingly enhanced number concentrations in the nucleation mode size range from 10 nm to 20 nm, regardless of the cloud cover being closed or broken (Fig. 11). In contrast, in the PNSD measured during the descent towards cloud level around 10:30 UTC, which we assume to be representative for the free troposphere, concentrations in the sub-20 nm size range were low. This PNSD should be considered semi-quantitative only, as it represents an average over an altitude range of more than 1000 m. 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

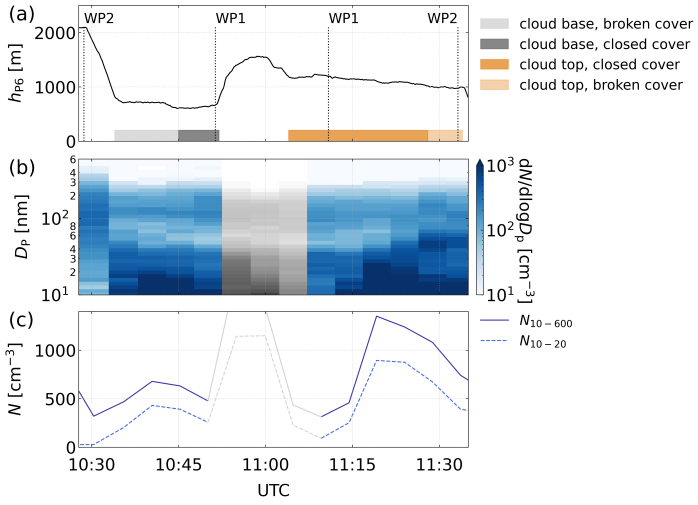


Figure 11. **(a)** P6 altitude during RF08 on 24 April 2024. **(b)** MPSS-measured PNSD. **(c)** Integral particle number concentrations in the total measured size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 600\ \mathrm{nm}$, solid line) and in the nucleation mode size range ($10\ \mathrm{nm} \leq D_\mathrm{p} \leq 20\ \mathrm{nm}$, dashed line), respectively. The data measured from 10:52 to 11:07 UTC are grayed out in panels (b) and (c) since erroneous measurements due to icing of the aerosol inlet and/or droplet shattering cannot be ruled out.

contributing approximately ~~65~~67 %, ~~59~~60 %, and 9 % to the total aerosol particle number concentrations at cloud top, cloud base, and in the free troposphere, respectively. A more detailed comparison between PNSDs measured at cloud top and at cloud base reveals an interesting effect (Fig. 12)~~reveals an interesting effect~~. The overall shapes of the distributions are qualitatively similar, with the cloud top and cloud base accumulation mode concentrations following each other rather closely, while the number of Aitken and nucleation mode particles is reduced by roughly a factor of two to three at cloud base, resulting in a more pronounced Hoppel-minimum for the PNSDs measured at cloud base. Even a small shift of the accumulation mode at cloud base to larger sizes can be seen, which could be due to particle growth inside cloud. ~~However, differences are too small to draw~~ 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. To indicate the absence of nucleation mode particles in the free troposphere, ~~also shown in Fig. 12 is~~ the PNSD measured during the descent from 2100 m towards cloud level ~~.~~is also shown in Fig. 12.

~~Cloud tops, due to the prevailing high humidities and high actinic fluxes, which enhance photochemically driven processes, as well as the presence of potential particle precursors released from evaporating droplets, have been reported to be favorable locations for NPF to take place (e.g., Hegg et al., 1990; Clarke et al., 1998; Weber et al., 2001; Keil and Wendisch, 2001; Wehner et al., 20 . Overall, this leads us to the hypothesis that NPF in the vicinity of the cloud top led to the observed elevated concentrations of newly formed particles at cloud top and cloud base, with the newly formed particles being simultaneously mixed down and~~

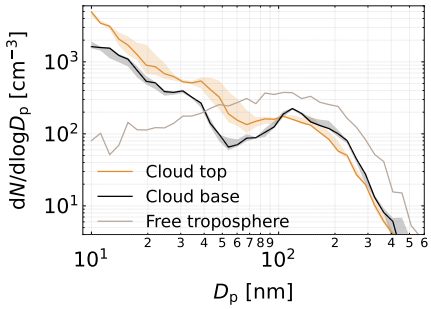


Figure 12. Median PNSDs measured at cloud top, cloud base, and in the free troposphere. The shaded areas represent the interquartile ranges.

~~cloud processed (wet scavenging and/or growth depending on their size). Such a process is conceivable for a marine ABL with the dynamics of radiatively cooled clouds (radiative cooling rate at cloud top $\approx -120\ \mathrm{K\,d^{-1}}$, Fig. S11, Lonardi et al. (2024)) and the related transport and mixing processes (Wood, 2012). 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.~~

Backward trajectory analysis of all air masses featuring newly formed particles in the vicinity of clouds

Fig.13 presents the results of the backward trajectory analysis carried out for all PNSD measurements featuring newly formed particles in the vicinity of clouds, which were observed during three different research flights on 23, 24 (case study), and 26 April.

Predominantly, the air masses approached from lower latitudes and moved northward, mainly in the last 48 hours prior to arrival (Fig. 13a). The trajectories traveled over the open sea in the last 18 to 60 hours and 90 to 100 hours (Fig. 13b), whereby they became warmer and picked up humidity (Fig. 13e-g). Since the moist air masses moved poleward, an increase of IVT_{north} is visible in the last 48 hours before arrival (Fig. 13h). Almost 75 % of the trajectories were in contact with the ABL during the last five days (Fig. 13i), presenting the opportunity for moisture and gaseous precursor uptake, potentially from the open ocean. In ~~89~~about 90 % of the air masses, clouds had formed within the previous five days (Fig. 13~~i~~j). The gradual decrease in potential temperature found in the last 24 hours prior to arrival (Fig. 13d) may result from the presence of these clouds, i.e., from cloud top radiative cooling.

~~Despite these similarities between the air mass histories, it is difficult to draw conclusions concerning the potential role of long-range transport in the observed events from~~ Cloud tops, due to the ~~data available. Nevertheless, in our view, our measurement data clearly suggest~~ prevailing high humidities and high actinic fluxes, which enhance photochemically driven processes, as well as the presence of potential particle precursors released from evaporating droplets, have been reported to be favorable locations for NPF to take place (e.g., Hegg et al., 1990; Clarke et al., 1998; Weber et al., 2001; Keil and Wendisch, 2001; Wehner . 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

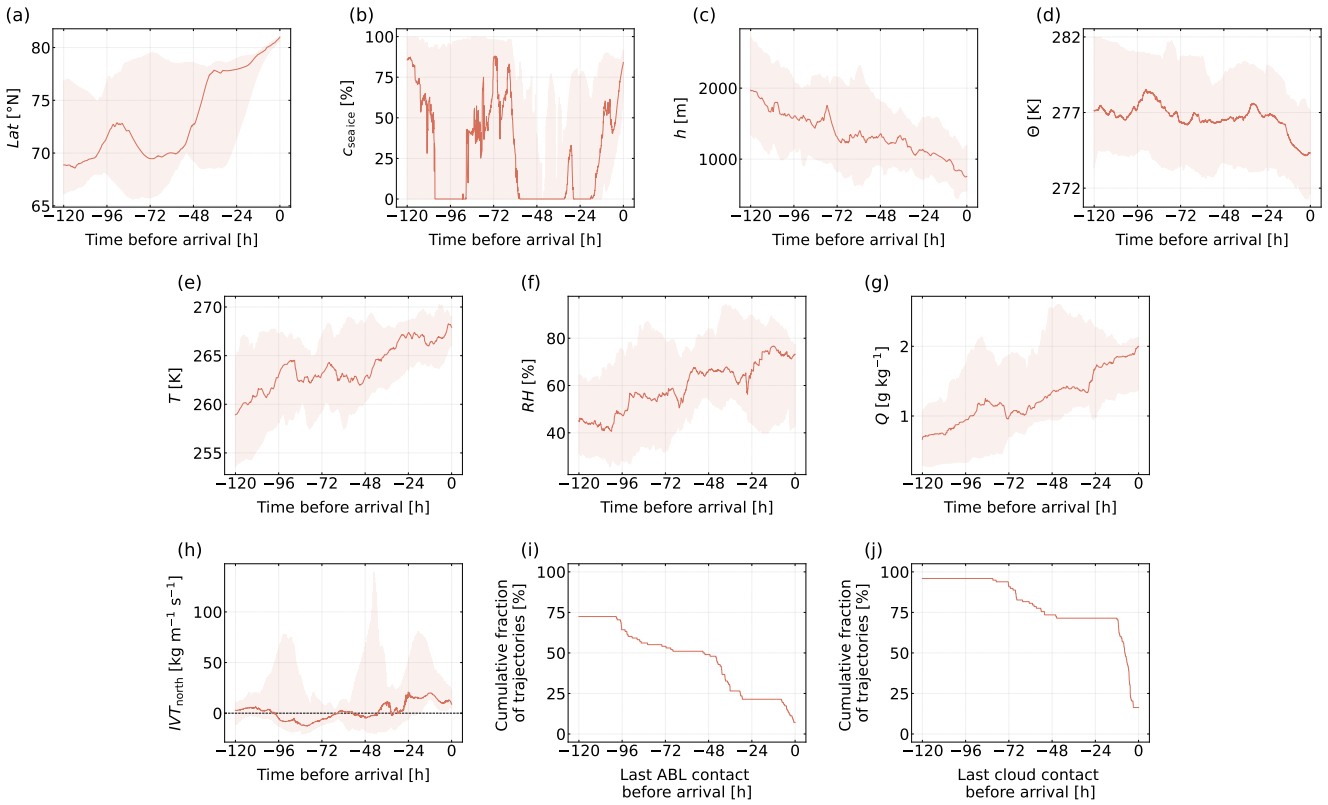


Figure 13. Meteorological parameters along backward trajectories linked to ~~for~~ newly formed particles detected in the vicinity of clouds. Shown, as a function of time prior to arrival, are median values (solid lines) and interquartile ranges (shaded areas) of **(a)** latitude Lat, **(b)** sea ice concentration c_{seaice}, **(c)** altitude h, **(d)** potential temperature Θ, **(e)** absolute temperature T, **(f)** relative humidity RH, **(g)** specific humidity Q, and **(h)** northward integrated water vapor transport IVT_{north}. The last two subplots show the cumulative fraction of the last contact of the trajectories with the **(i)** ABL and **(j)** clouds. Note that if the cumulative fraction does not increase to 100 % at $t = -120\,\mathrm{h}$, this indicates that a fraction of the trajectories was not in contact with the ABL/clouds during the last five days (the maximum time span the calculated trajectories cover).

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).

Overall, this leads us to the hypothesis that NPF in the vicinity of ~~clouds was observed~~ the cloud top led to the observed elevated concentrations of newly formed particles at cloud top and cloud base, with the ~~region near clouds providing a favorable environment for NPF to take place.~~ newly formed particles being simultaneously mixed down and cloud processed (wet scavenging and/or growth depending on their size). Such a process is conceivable for a marine ABL with the dynamics of radiatively cooled clouds (radiative cooling rate at cloud top $\approx -120\,\mathrm{K\,d^{-1}}$, Fig. S14, Lonardi et al. (2024)) and the related

transport and mixing processes (Wood, 2012). Considering the abundance of Arctic stratiform mixed-phase clouds (e.g., Shupe et al., 2013), NPF in the vicinity of such clouds may play an important role in the Arctic aerosol particle and CCN budgets.

4 Conclusions

In this study, we discussed the frequent occurrence of newly formed aerosol particles in the Arctic troposphere as observed during ~~an~~ the airborne measurement campaign ~~utilizing the Polar 6 research aircraft~~BACSAM II, which was conducted from 5 April 2024 to 2 May ~~2024.~~ 2024 utilizing the Polar 6 aircraft (Wesche et al., 2016). The Polar 6 was instrumented with dedicated meteorological and aerosol instrumentation collecting data over and around the Svalbard Archipelago, over the Fram Strait, and in the vicinity of Villum Research Station (Station Nord) in Greenland. We observed newly formed particles during all eleven research flights at various locations (approximately between 76°N to 85°N and 15°W to 26°E). Newly formed particles were encountered in an altitude range from about 60 m to 3900 m above sea level, i.e., throughout the flight altitude range ~~covered by our aircraft~~investigated during the campaign. Specifically, these particles were detected in the free troposphere, inside the atmospheric boundary layer (ABL) over sea ice, ~~and~~ 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.

Newly formed particles in the free troposphere were encountered across large geographical areas (with individual events spanning distances of hundreds of kilometers) and at different altitudes. Our respective analyses suggest a possibly important role of meridional transport in combination with cloud processing ~~in Arctic~~ and precipitation for free tropospheric new particle formation (NPF)~~as~~. Such a scenario could be encountered during, e.g., warm-air intrusions which may deliver the precursor gases to atmospheric conditions (high altitudes, intense solar radiation, low temperatures, small pre-existing particle surface area) favorable for NPF. With respect to the newly formed particles detected inside the ABL over sea ice, we ~~conclude~~ hypothesize that these particles are ~~presumably~~ the result of regional NPF events induced by precursor gases (conceivably iodine components) originating from the sea ice, leads, or snow present in the measurement region. However, contributions of higher-latitude Arctic (precursor) sources cannot be ruled out. Concerning newly formed particles observed at both cloud tops and cloud bases, we found ~~clear~~ indications that these particles are ~~very likely~~ 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. The presence of newly formed particles inside the ABL over ocean with sea ice floes could partly be related to a phytoplankton bloom which occurred in the respective target region on the day of the research flight. But ultimately, the source of precursors which could contribute to NPF could not be identified.

Taken together, our results show that newly formed particles were frequently present and very likely produced in the both the Arctic free troposphere and the Arctic ABL during our measurements in spring ~~2024, while prior observations predominantly suggested that NPF inside the ABL is the most common source of secondary aerosol particles in the Arctic.~~ 2024. In particular, for the first time, our study presents observational results which indicate that NPF in the Arctic free troposphere ~~could be a geographically widespread phenomenon, and that~~ occurred over distances of several hundreds of kilometers. In this context, a

possible mechanism leading to Arctic free tropospheric NPF ~~is connected to~~ could be associated with warm-air intrusions. ~~This~~ A similar transport pathway of precursor gases and/or newly formed particles from lower latitudes to the Arctic free troposphere was previously suggested in a modeling study by Price et al. (2023), but to date had not been substantiated by analysis of in situ-measured data. We present observational evidence which ~~supports the findings of Price et al. (2023) and emphasizes the importance of free tropospheric sources of newly formed particles~~likewise shows that meridional transport can lead to NPF in the Arctic free troposphere.

Further modeling studies are required to assess the impact of newly formed particles occurring over ~~large geographical areas~~ distances of hundreds of kilometers and a wide range of altitudes inside and above the Arctic ABL on the Arctic aerosol and cloud condensation nuclei (CCN) budgets~~, and ultimately on the Arctic climate~~. In particular, because relatively small, sub-100 nm particles readily act as CCN in the clean Arctic atmosphere, the ~~geographically-extended, frequent~~ occurrence of newly formed particles in the free troposphere and the ABL might exert an important influence on Arctic aerosol-cloud-climate interactions. Future observational campaigns would profit from trace gas measurements and fast, size-resolved measurements of particles with diameters of less than 10 nm, e.g., with multiple condensation particle counters featuring different lower cut-off diameters. Such measurements would facilitate the identification of the relevant precursor gases and the precise nucleation region. Importantly, prospective studies should include vertical measurements and target both the Arctic ABL and free troposphere. Here, the use of drop sondes to provide high-resolution profiles of the vertical structure of the atmosphere would be beneficial.

Finally, in a rapidly warming Arctic, where the sea ice continues to retreat, more clouds might be present in the future, and the occurrence of warm-air intrusions is expected to increase, NPF frequency and location will likely be affected by these changes. Especially NPF in the free troposphere, possibly linked to warm-air intrusions, and at the tops of Arctic stratiform clouds, might significantly impact current and future Arctic aerosol and CCN budgets due to its potentially large geographical extent and frequent occurrence, and should therefore be considered in atmospheric models for the prediction of future Arctic weather and climate.

Data availability. Master tracks are available on PANGAEA for all research flights of the BACSAM II campaign (Jurányi et al., 2024). Backward trajectory data are available on PANGAEA for all research flights of the BACSAM II campaign (Kirbus and Wendisch, 2024). PNSD data from all research flights of the BACSAM II campaign are available on PANGAEA (Simon et al., 2026). Sea ice data were retrieved from www.seaice.uni-bremen.de (Spreen et al., 2008). MODIS-TERRA satellite images were retrieved from https://wvs.earthdata.nasa.gov. This study has been conducted using E.U. Copernicus Marine Service Information; https://www.doi.org/10.48670/moi-00281.

Author contributions. DJS participated in the data collection and flight planning during the BACSAM II campaign, analyzed and interpreted the data, and wrote the manuscript. JS participated in the data collection and flight planning, discussed the data, and contributed to the writing. JH participated in the data collection and flight planning, and processed the position, nose boom, and humidity data. BK performed the trajectory calculations and supported their analysis. JM participated in the data collection, flight planning, and calculated the radiative

heating/cooling rates. ZJ, MH, BW, LK,AMJ, and MW participated in the flight planning, and the data collection and related discussions. KW was responsible for the development of the MPSS system. AH and ZJ functioned as PIs during the campaign. AH, ZJ, SH, GCR, MW, and MLP contributed to the discussion of the results. FS conceived the study, functioned as PI during the campaign, and contributed to data interpretation and writing. All authors read and edited the manuscript.

Competing interests. The authors declare no competing interests.

Acknowledgements. We gratefully acknowledge the funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – Project Number 268020496 – TRR 172, within the framework of the Transregional Collaborative Research Center "ArctiC Amplification: Climate Relevant Atmospheric and SurfaCe Processes, and Feedback Mechanisms $(AC)^3$", as well as the support of the Alfred-Wegener-Institute, Helmholtz Centre for Polar and Marine Research through grant number AWI_PA_02172. The authors used a large language model (ChatGPT by OpenAI) solely to support coding tasks; all scientific reasoning, methodological decisions, analyses, and conclusions were developed independently by the authors.

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Supporting information to "Frequent occurrence of newly formed aerosol particles over wide geographical areas in the Arctic free troposphere and atmospheric boundary layer"

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S1 Research flights during the aircraft measurement campaign

Fig. S1 shows the flight tracks of all eleven research flights (RFs) conducted with the Polar 6 (P6) aircraft during the measurement campaign "Boundary layer and Aerosol and Cloud Study in the Arctic, based on aircraft and T-Bird Measurements II" (BACSAM II).

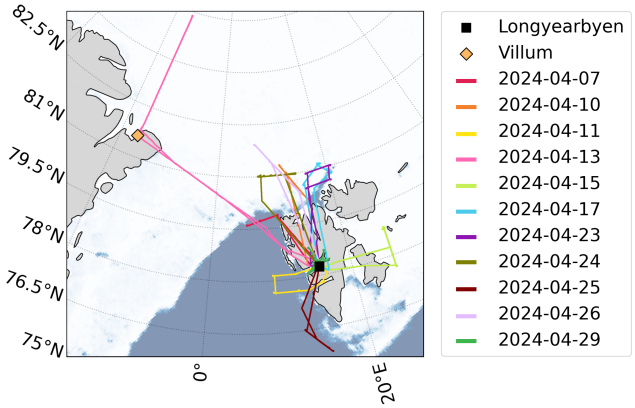


Figure S1. Flight tracks of all research flights during BACSAM II. The sea ice concentration is shown exemplary for 15 April 2024. The corresponding sea ice data were retrieved from www.seaice.uni-bremen.de (Spreen et al., 2008).

S2 ~~Detection and categorization of newly formed particles~~Correction for particle losses

S2.1 ~~Detection of newly formed particles~~

~~As outlined in the main text (Sect. 2.2), we fitted log-normal functions featuring one, two, or three modes~~ To characterize particle losses in the sampling line from the aerosol inlet of the Polar 6 aircraft to the mobility particle size ~~spectrometer-measured (MPSS-measured)particle number size distribution (PNSD) data. If the fitting procedure returned a mode featuring a maximum below 10 nm, i.e., below the lower size limit of the MPSS, we consider the measurement to indicate the presence of newly formed particles. The detection of newly formed particles was verified by visual inspection of the fitted PNSDs. As outlined in~~ 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, ~~MPSS measurements carried out during ascents and descents were excluded from the quantitative analysis. Fig. **??** shows examples of three fitted PNSDs, with the data displayed in Fig. **??**a and Fig. **??**c indicating the presence of newly formed particles, and the data~~ 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. S2. 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. ~~**??**b the absence of newly formed particles. Examples of MPSS-measured PNSDs~~S2 as well. Here, the respective parameters are set to $p_{10} = 678\,\mathrm{hPa}$, ~~showing nucleation and/or Aitken~~$T_{10} = 286\,\mathrm{K}$, ~~and accumulation modes. The PNSDs shown in the panels (a) and (c) are considered as indicative~~ $p_{90} = 905\,\mathrm{hPa}$, $T_{90} = 296\,\mathrm{K}$ for ~~newly formed particles~~$tas \approx 90\,\mathrm{m\,s^{-1}}$, and $p_{10} = 862\,\mathrm{hPa}$, ~~while the measurement displayed~~ $T_{10} = 293\,\mathrm{K}$, $p_{90} = 1030\,\mathrm{hPa}$, $T_\mathrm{med} = 300\,\mathrm{K}$ for $tas \approx 60\,\mathrm{m\,s^{-1}}$.

S2.1 ~~Categorization of newly formed particles~~

~~The~~

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. S2). 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. S2). Accordingly, we correct the MPSS-measured ~~PNSDs indicative of newly formed particles~~

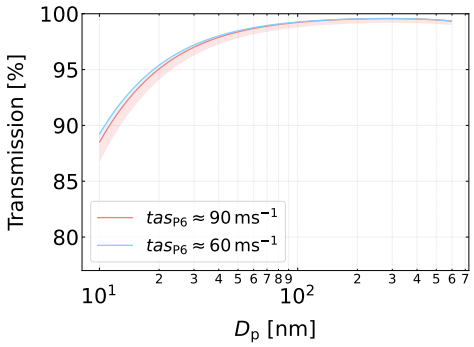


Figure S2. Particle size-dependent transmission efficiencies in ~~panel (b) shows no indication~~ the sampling tubing from the aerosol inlet to the MPSS for ~~newly formed particles~~the two typical true airspeeds of the Polar 6 aircraft during the campaign.

~~were categorized according to three environmental conditions for which they were mainly observed, i.e., newly formed particles detected in the free troposphere (Sect. 3.2 in the main text)~~ particle number size distribution (PNSD) data for particles losses in the sampling tubing by size-dependent correction factors ($c(D_p) = 1/\eta(D_p)$) 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.

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 ~~atmospheric boundary layer (ABL) over sea ice (Sect. 3.3), and~~ (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 ~~(Sect. 3.4) . Fig. ?? shows the median PNSDs and corresponding interquartile ranges of all MPSS measurementsper category. For convenience, the PNSDs are shown on both a logarithmic (upper panel) and linear axis (lower panel).~~ 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.

~~For approximately 14 % of all PNSDs featuring~~ 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, approximately 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 ~~, the prevailing conditions could not unambiguously be identified.These include flight sections during which clouds were present, but in contrast to the~~ on this day (≈ 10 minutes) are assigned to the unclassified category.

In addition, the relative frequencies of occurrence of newly formed particles ~~classified as "in the vicinity of clouds", we did not fly directly at~~ 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 ~~base, respectively. Contained in the 14 % of unclassified cases are also sections at rather low altitudes (60 - 300 m, see Fig. 2b in the main text) over ocean, which were at least partly carried out inside the marine ABL . Here, either clouds were present as well, or sea icefloes were visible in addition to open sea areas, preventing the clear identification of the potential precursor source (open sea or sea ice). Consequently, we cannot rule out that the open ocean acted as source of precursor gases (e.g., dimethyl sulfide (DMS), Willis et al., 2018) leading to~~ 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 ~~in some of the regions targeted during our campaign, but , in contrast to the three other cases investigated, no clear observations of newly formed particles over open ocean were made. Median PNSD of all cases of newly formed particles associated with one of the three environments described in the main text and discussed in detail in the respective sections 3.2 (free troposphere), 3.3 (ABL over sea ice), and 3.4 (cloud vicinity). The shaded areas represent the interquartile ranges.~~ 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.

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.

S4 Thermodynamic conditions ~~prevailing~~ during the occurrence of newly formed particles

In Fig. S3, relative frequencies of occurrence of measured temperature and relative humidity pairs (T-RH pairs) are plotted in a temperature vs. relative humidity plane. The relative frequencies were ~~calculated~~ computed considering the number of measured T-RH pairs during the presence of newly formed particles (Fig. S3a, ~~37452~~ 3686703 pairs) and considering the number of T-RH pairs measured during the absence of newly formed particle (Fig. S3b, ~~116891~~ 9094434 pairs). The histograms shown in Fig. S3~~c-c~~ c-f were calculated by ~~normalizing~~ including the T-RH data pairs recorded only during observations of newly formed particles in the free troposphere (Fig. S3c), only during observations of newly formed particles in the ABL over sea ice (Fig. S3d), ~~and~~ only during observations of newly formed particles in the vicinity of clouds (Fig. S3e), and only during observations of newly formed particles in the ABL over ocean with sea ice floes (Fig. S3f). These data are then normalized by the data recorded during all observations of newly formed particles combined. That is, the normalization is the same as for the histogram depicted in Fig. S3a. The dotted horizontal and vertical lines separating the subplot into four quadrants in Fig. S3 are plotted to guide the eyes.

From Fig. S3, a clear pattern becomes visible. The data show three main clusters in the relative frequency distribution of temperature and humidity conditions during observations of newly formed particles (Fig. S3a), while the frequency distribution of T-RH pairs measured during the absence of newly formed particles is broader, with fewer pronounced features (Fig. S3b). The peaks in Fig. S3a, i.e., in the T-RH distribution measured during observations of newly formed particles, correspond to the different environmental scenarios under which newly formed particles were detected. Newly formed particles in the free troposphere were predominantly associated with cold and dry conditions (lower left quadrant in Fig. S3a, Fig. S3c). Newly formed particles in the ABL over sea ice occurred during cold and humid conditions (lower right quadrant in Fig. S3a, Fig. S3d). For newly formed particles detected in the vicinity of clouds, relatively warm and humid conditions prevailed (upper right quadrant in Fig. S3a, Fig. S3e). Similarly, newly formed particles inside the ABL over ocean with sea ice floes occurred

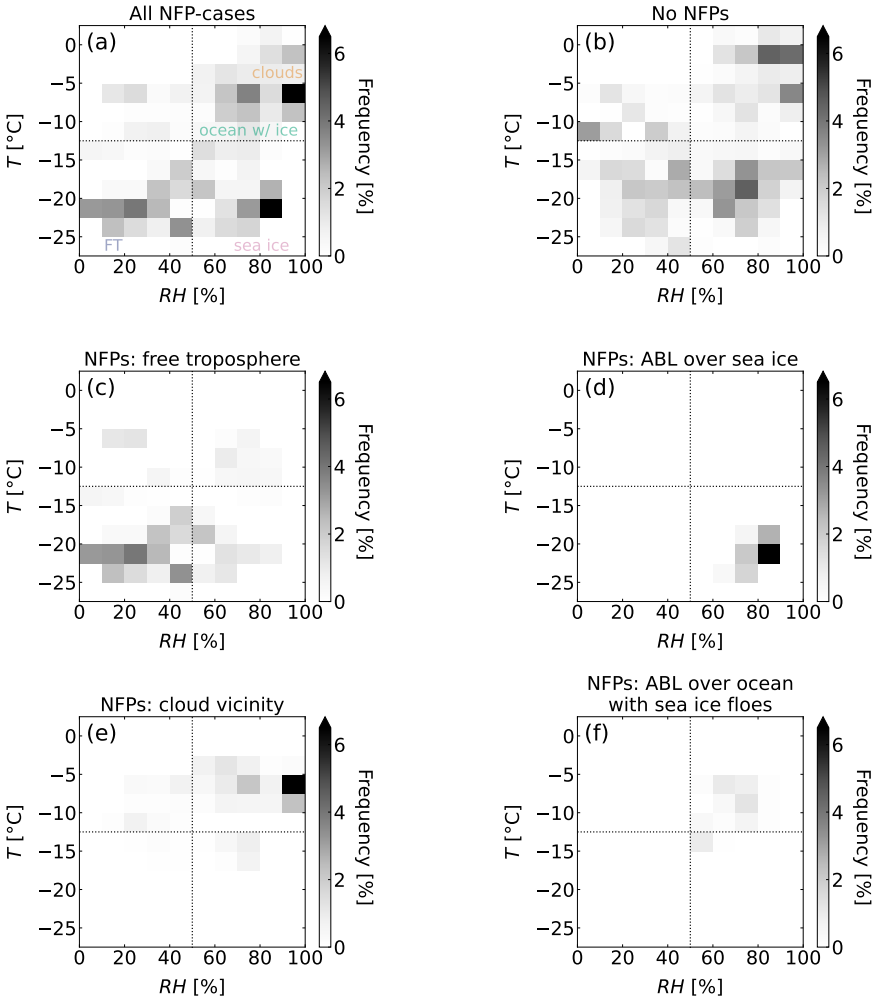


Figure S3. Temperature and relative humidity conditions during **(a)** newly formed particle-cases (NFP-cases) and **(b)** non-NFP-cases. The labels "FT", "sea ice", ~~and~~ "clouds", and "ocean w/ ice" in panel (a) denote that within the corresponding quadrant, the measured temperature and relative humidity values are predominantly associated with newly formed particles observed in the free troposphere, the ABL over sea ice, ~~and~~ cloud vicinity, and the ABL over ocean with sea ice floes, respectively. The second and third row ~~depicts~~ depict temperature and relative humidity during the occurrence of newly formed particles **(c)** in the free troposphere, **(d)** the ABL over sea ice, **(e)**, ~~and~~ the vicinity of clouds, **(f)** and the ABL over ocean with sea ice floes.

during relatively warm, but less humid conditions compared to the observations in cloud vicinity (upper right quadrant in Fig. S3a, Fig. S3f). As the latter cases comprise only about 40 minutes of measurements (6 % of all observations of newly formed particles), the corresponding peaks in Fig. S3f are not strongly pronounced.

As mentioned in Sect. 3.1 in the main text and discussed comprehensively in supporting information (SI) Sect. ??S5, we found no clear indications for newly formed particles inside the ABL over open, completely ice-free ocean during our campaign. T-

RH data measured during such observations would likely also feature warm and humid conditions, similar to the data recorded during observations of newly formed particles in the vicinity of clouds ~~,~~ and in the ABL over ocean with sea ice floes. Altogether, these results show that newly formed particles observed during BACSAM II ~~seem to be~~ are associated with distinct thermodynamic conditions, which correspond to the different atmospheric scenarios for which newly formed particles were encountered~~, i.e.,~~. They include free tropospheric newly formed particles, newly formed particles inside the ABL over sea ice ~~, and~~ and over ocean with sea ice floes, as well as newly formed particles in the vicinity of clouds. These data provide important information on boundary conditions for laboratory experiments and modeling studies, both of which could facilitate the identification of nucleation mechanisms that might have lead to the presence of newly formed particles, and, furthermore, to assess the broader atmospheric (climate) impact of newly formed particles occurring throughout the lower Arctic troposphere.

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. S4a). The air masses probed during the legs inside the ABL originated in sea ice-covered regions north and east of Svalbard (Fig. S4b, 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. S4b, c). While mostly clear-sky conditions prevailed, a few scattered clouds were present in the eastern parts of the target region as well (Fig. S4a).

On 25 April, measurements were carried out in a target region southwest of Spitsbergen over open, completely ice-free ocean (Fig. S4d). On this day, air masses approached from southern latitudes (Fig. S4e, f). For the air masses probed inside the ABL (Fig. S4f), 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. S4f). 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. S4d).

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. S5a, see Fig. S4a for the location of the WPs). One leg was conducted at $\approx$ 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.

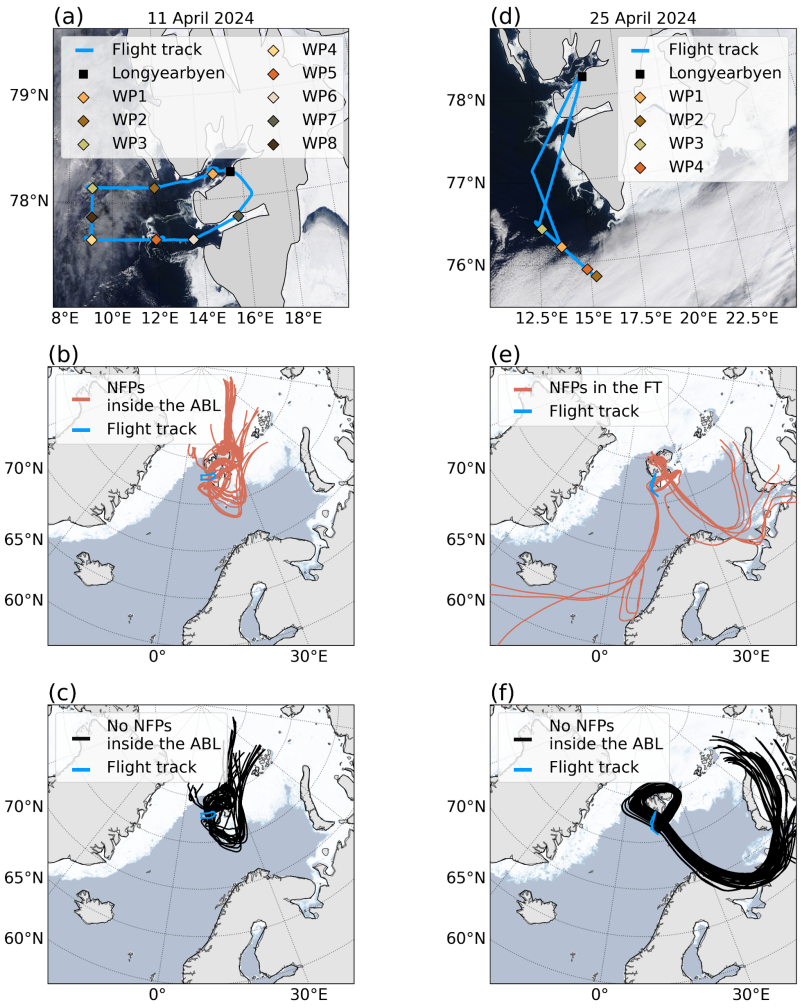


Figure S4. **(a)** Flight track of RF03 on 11 April 2024. **(b)** Five-day backward trajectories associated with newly formed particles (NFPs) inside the ABL on 11 April. **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles inside the ABL on 11 April. **(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).

S5a, see Fig. S4a 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. S5a, 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. S5a, 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. S4b, Fig. S5a).

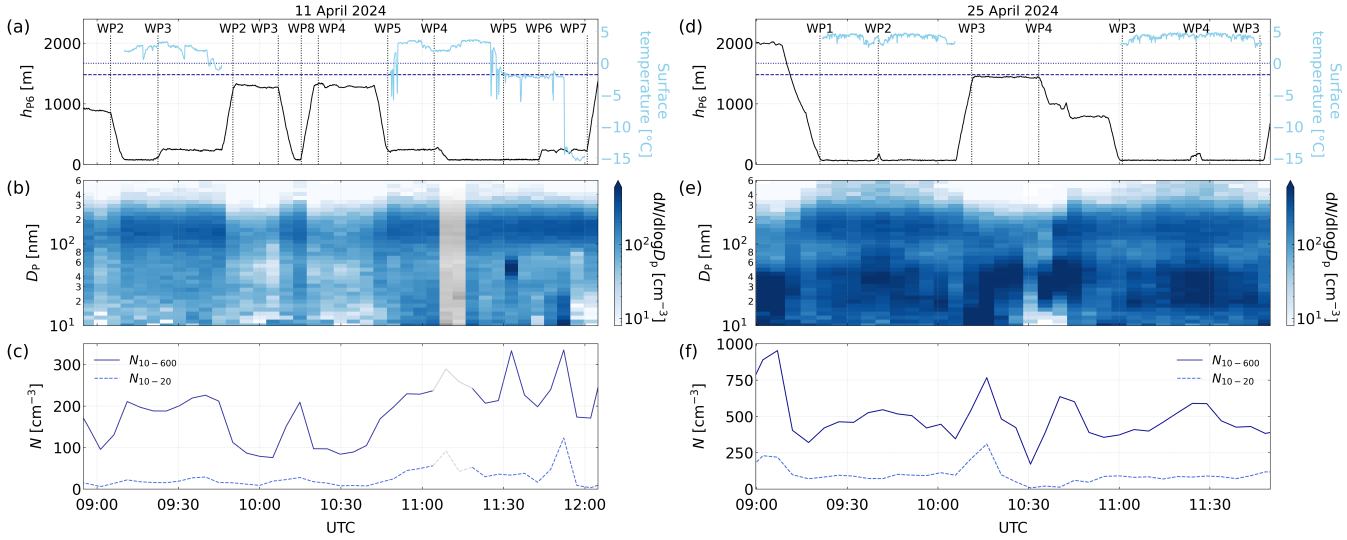


Figure S5. **(a)** P6 altitude during RF03 on 11 April 2024 (black line, left y-axis) as well as surface temperature (blue line, right y-axis). The dashed/dotted blue horizontal lines illustrate a surface temperature of 0°C and the freezing temperature of (salty) Arctic ocean water of −1.8°C. **(b)** 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 ($10\ \mathrm{nm} \leq D_p \leq 600\ \mathrm{nm}$, solid line) and in the nucleation mode size range ($10\ \mathrm{nm} \leq D_p \leq 20\ \mathrm{nm}$, 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 ($10\ \mathrm{nm} \leq D_p \leq 600\ \mathrm{nm}$, solid line) and in the nucleation mode size range ($10\ \mathrm{nm} \leq D_p \leq 20\ \mathrm{nm}$, dashed line), respectively. Note the different y-axis between the subplots (c) and (f).

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. S5b, c). In the northern section, no newly formed particles during the legs inside the ABL over completely ice-free ocean were present (Fig. S5b, 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 ($N_{10-20} \approx 25\,\mathrm{cm}^{-3}$, $\approx 10\,\%$ 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. S5b). 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).

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. S5c). 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. S5b, 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. S5b). The median PNSDs for the legs inside the ABL are shown in Fig. S6 and will be discussed below.

On 25 April, four legs at altitudes of $\approx$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. S5d, left y-axis), i.e., with no sea ice present at all, as shown by a sea surface temperature of about 4°C (Fig. S5d, right y-axis). Additionally, one leg was flown at $\approx$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 $\approx$1000 m to $\approx$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. S5e, 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. S4e). 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. S6. 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. S6) 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 spee
. 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. S6). In contrast, PNSDs measured inside the ABL over water with sea ice floes indicated the presence of newly formed particles (red distributions in Fig. S6).

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.

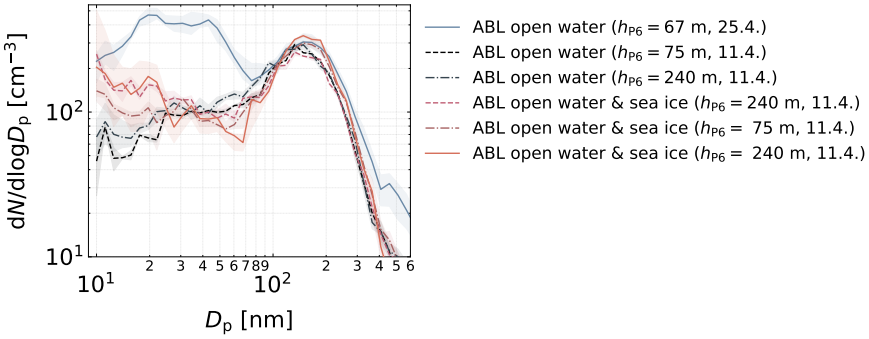


Figure S6. Median particle number size distributions during legs at constant altitude inside the ABL over completely ice-free open water (on 11 and 25 April, black and blue curves) and over ocean with both sea ice floes and open water areas present (on 11 April, red curves). The latter distributions 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.

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. S7a) where both open water and sea ice were present (Fig. S4a, S5a), 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. S7a).

Fig. S7b 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. S7b-d, f-h. As mentioned above, easterly winds prevailed in both the northern

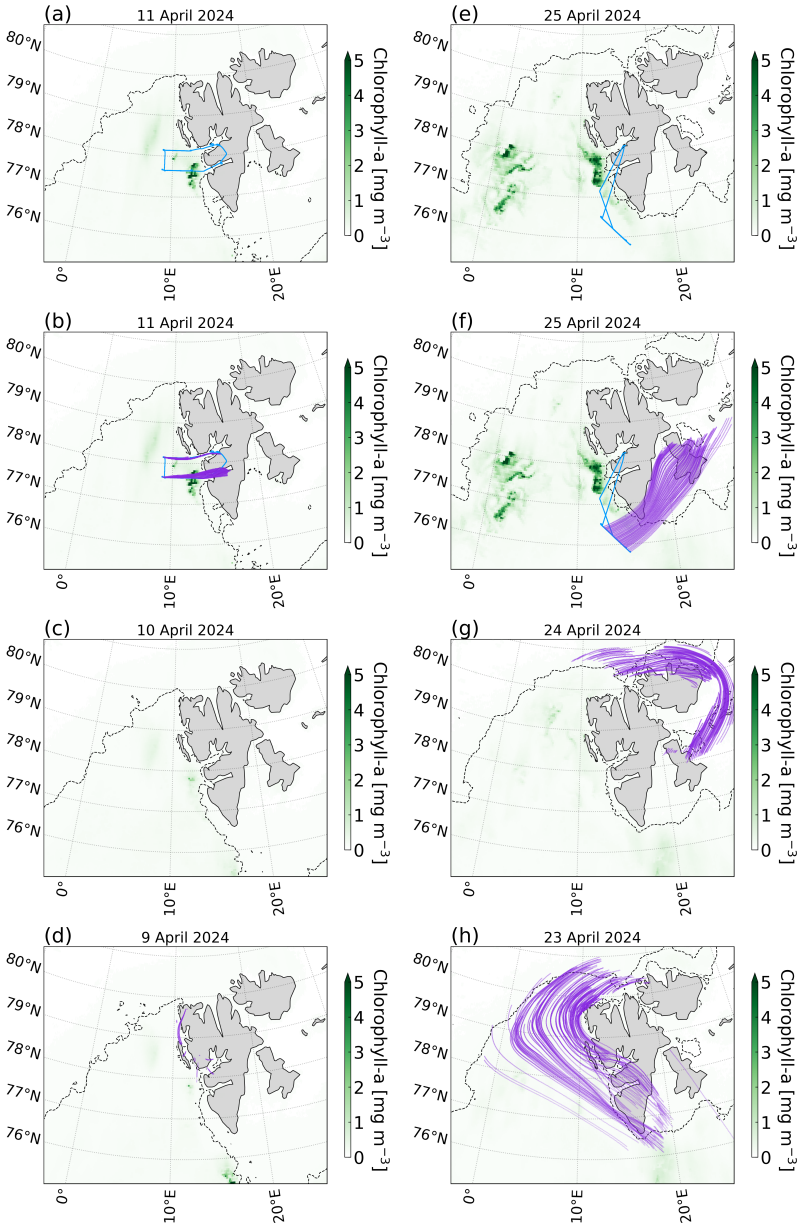


Figure S7. Flight track (blue), backward trajectories inside the ABL (violet), and chlorophyll-a mass concentrations (green) based on Copernicus-GlobColour data (Gohin et al., 2002; Hu et al., 2012) for the research flights featuring measurements inside the ABL over ocean on 11 and 25 April. The left 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).

and the southern section (Fig. S7b). 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. S7b). 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. S7c) and overwhelmingly outside of the ABL (9 April, Fig. S7d), 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. S7e). 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. S7f-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. S5a-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. S7b). 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).

S6 New and recently formed particles in the free troposphere

Fig. S8 shows the median PNSD and corresponding interquartile range of all PNSD-measurements indicative of newly formed particles during the transit to Villum research station in the free troposphere (13 April 2024, RF04, Sect. 3.2 in the main text). These data were recorded during the time period 6:23 to 7:07 UTC and 7:17 to 7:40 UTC.

~~Median PNSD of measurements indicative of newly formed particles (NFPs) and recently formed particles (RPFs, solid lines) observed on 13 April 2024 during the transit from Longyearbyen to Villum Research Station (case study in the free troposphere). The shaded areas represent the interquartile ranges.~~ Two PNSDs measured in the time period from 7:07 to 7:17 UTC peaked at about 18 nm and showed no indications concerning a mode featuring a maximum below 10 nm (Fig. S8). According to our criteria defined ~~above~~in Sect. 2.2 in the main text, these PNSDs are not considered as indicative for newly

formed particles. We rather interpret them as steming from "recent particle formation" (RPF), since the small particle sizes still suggest that these particles must originate from secondary particle production (e.g., Kulmala et al., 2004). Such recently formed particles are excluded from further analysis and we here focus solely on newly formed particles. Investigating recently formed particles in ~~further~~ more detail is beyond the scope of this paper.

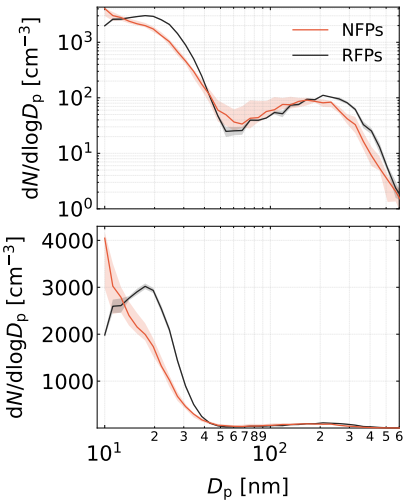


Figure S8. Median PNSD of measurements indicative of newly formed particles (NFPs) and recently formed particles (RFPs, solid lines) observed on 13 April 2024 during the transit from Longyearbyen to Villum Research Station (case study on newly formed particles in the free troposphere). The shaded areas represent the interquartile ranges.

S7 Newly formed particles in the atmospheric boundary layer over sea ice

S7.1 Sea ice conditions

To illustrate the sea ice conditions prevailing in the target region east of Svalbard between ~~waypoint 1 (~~WP1 ~~)~~ and WP3 on 15 April 2024 (RF05, Sect. 3.3 in the main text), Fig. S9 displays examples of fisheye camera images facing downward from the P6 aircraft during the legs inside the ABL. Leads and cracks in the sea ice are visible.

S7.2 Vertical profile

From the inversion observed in the vertical profiles of potential temperature (Θ) between WP1 and WP3, we determine the ABL height (Fig. S10). The solid line in Fig. S10 shows the mean Θ-profile averaged over the four ascents/descents at WP1, WP2, and WP3 (see Fig.~~7a~~ 8a in the main text). The shaded area represents the standard deviation and thus the variability between the individual ascents/descents. The data have been grouped into altitude bins with a width of 20 m before averaging. The markers with errorbars show mean values and standard deviations derived from the measurements during the horizontal legs. The

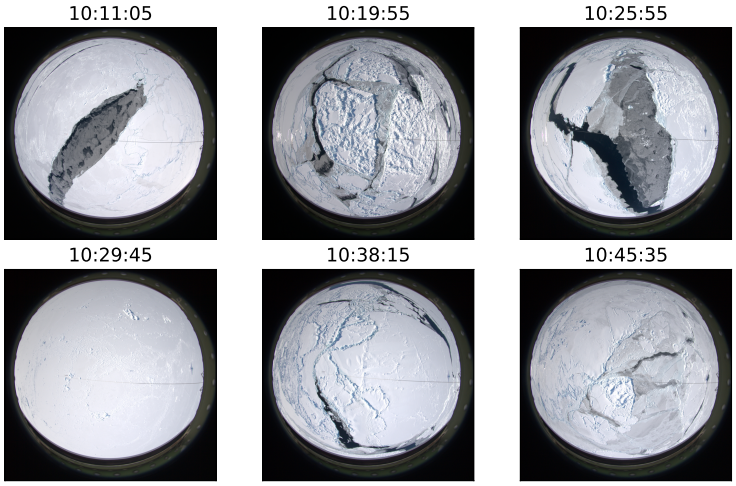


Figure S9. Sea ice conditions during RF05 on 15 April 2024 (case study in the ABL over sea ice). The title denotes the time in UTC.

horizontal dashed lines represent the average aircraft altitude during these low-level legs (at $\approx 70\,\mathrm{m}$ and $\approx 155\,\mathrm{m}$) close to the surface inside the ABL ($h_{\mathrm{P6,\,inside\,BL}}$) and the high-level leg above the ABL in the free troposphere ($h_{\mathrm{P6,\,outside\,BL}} \approx 850\,\mathrm{m}$). In Fig. S10, an inversion at an altitude of about ~~170~~ 190 m can be identified for the target region.

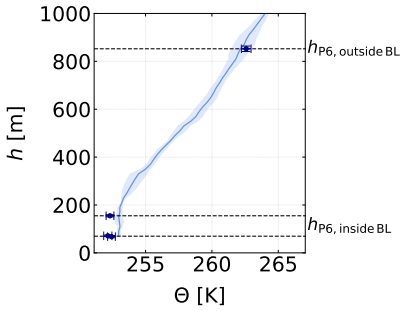


Figure S10. Mean vertical profile of potential temperature for the case study in the ABL over sea ice on 15 April 2024, derived from the ascents/descents (solid line). The shaded area represents the standard deviation. The markers with errorbars show mean values and standard deviations derived from the measurements during the horizontal legs. The horizontal dashed lines illustrate the average aircraft altitude during these legs.

S7.3 Particle number size distributions

Fig. S11 shows the median PNSDs measured during the three consecutive legs carried out in the same area and at constant altitude between WP1 and WP3 (see Fig. ~~7a~~ 8a in the main text) inside the ABL over sea ice. ABL 1 refers to the first leg flown inside the ABL at an altitude of $\approx 70\,\mathrm{m}$ (10:15 to 10:47 UTC), ABL 2 to the second leg at $\approx 155\,\mathrm{m}$ (10:58 to 11:25 UTC), and

ABL 3 to the third leg, again at ≈ 70 m (12:13 to 12:43 UTC). The decrease with time of the number concentration in the size range below 20 nm is clearly visible.

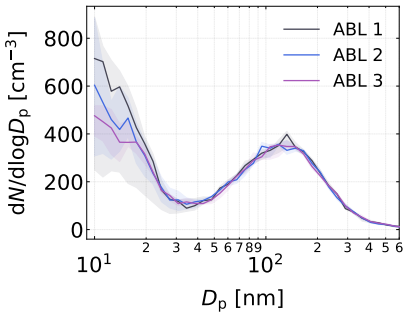


Figure S11. Median PNSDs (solid lines) and corresponding interquartile ranges (shaded areas) for the legs at ≈ 70 m (ABL 1 and ABL 3) and ≈ 155 m (ABL 2) inside the ABL over sea ice on 15 April 2024.

S8 Newly formed particles in the vicinity of clouds

S8.1 Sea ice conditions

To illustrate the sea ice conditions prevailing in the target region north of Svalbard between WP1 and WP2 on 24 April 2024 (RF08, Sect. 3.4 in the main text), Fig. S12 displays examples of fisheye camera images facing downward from the P6 aircraft during the leg at cloud base. Leads and cracks in the sea ice are visible.

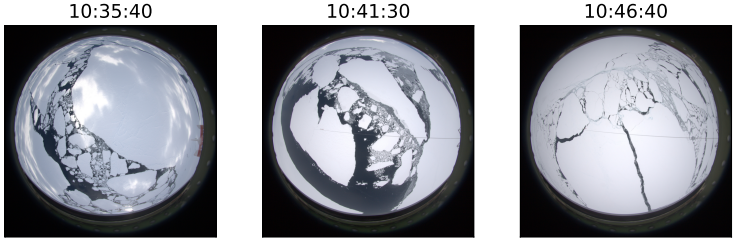


Figure S12. Sea ice conditions during RF08 on 24 April 2024 (case study in the vicinity of clouds). The title denotes the time in UTC.

S8.2 Vertical profiles

For the case study on newly formed particles in the vicinity of clouds, the ascent through cloud (10:52 to 10:58 UTC, see Fig. ~~10a~~ 11a in the main text) is excluded from analysis with respect to the aerosol particle measurements, because erroneous measurements due to icing of the aerosol inlet and/or droplet shattering cannot be ruled out (Weber et al., 1998). Nevertheless, temperature and pressure data recorded during the ascent can with caution be used to determine vertical profiles of potential

temperature to investigate the stratification in the target region. In Fig. S13, we show the potential temperature derived from these data collected during the ascent through cloud (see Fig. ~~10a~~ 11a in the main text). Directly in the target region between WP1 and WP2 (see Fig. ~~9a~~ 10a in the main text), no vertical profiling down to the lowest possible flight altitude was carried out. Therefore, in addition, the potential temperature derived from a vertical profile down to $\approx 80\,\mathrm{m}$, which was conducted after passing WP2 and while returning ~~towards~~ toward the sea ice edge (see Fig. ~~9~~10(a) in the main text), is depicted in Fig. S13. Here, the solid line represents the mean, averaged over ascent and descent, and the shaded area the corresponding standard deviation. Additionally, we show the potential temperature profile in the target region between WP1 and WP2 based on Copernicus Arctic Regional Reanalysis (CARRA) data. Cloud top and base, illustrated by the shaded gray area representing the cloud in Fig. S13, were determined from the profile of potential temperature and from in situ video data. Both CARRA and in situ data show an inversion below the cloud, which suggests that in the area probed between WP1 and WP2, the clouds were decoupled from the surface.

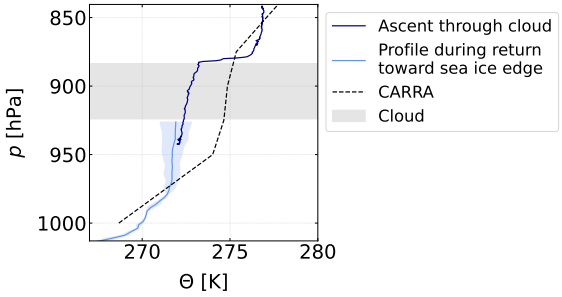


Figure S13. Vertical profile of potential temperature for the case study in the vicinity of clouds on 24 April 2024, derived from in situ data collected during ascents/descents (solid lines) as well as based on CARRA reanalysis data (dashed line).

S8.3 Cloud top radiative cooling

We further utilize the ascent through the cloud to determine the radiative heating/cooling rates in an atmospheric layer between two flight altitudes, with $z_{\mathrm{bottom/top}}$ denoting the altitudes at the bottom/top of this layer (Lonardi et al., 2024). Following Lonardi et al. (2024), we calculate the heating/cooling rates as

$$\zeta = \frac{1}{\rho c_{\mathrm{p}}} \frac{F_{\mathrm{net}}(z_{\mathrm{top}}) - F_{\mathrm{net}}(z_{\mathrm{bottom}})}{z_{\mathrm{top}} - z_{\mathrm{bottom}}}, \tag{1}$$

where ρ and c_{p} are density and specific heat capacity at constant pressure of air, and $F_{\mathrm{net}}(z)$ the net thermal irradiance at flight altitude z. Positive/negative values in ζ indicate radiative heating/cooling (Lonardi et al., 2024). The heating/cooling rates are plotted in Fig. S14, which shows a sharp minium at about 1150 m, i.e., radiative cooling at the cloud top.

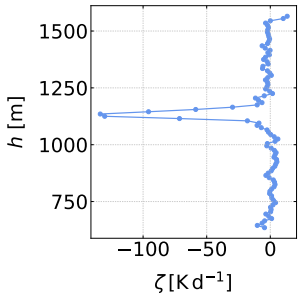


Figure S14. Radiative heating/cooling rates for the case study in the vicinity of clouds on 24 April 2024, determined from the ascent through cloud.

Data availability. Master tracks are available on PANGAEA for all research flights of the BACSAM II campaign (Jurányi et al., 2024). Backward trajectory data are available on PANGAEA for all research flights of the BACSAM II campaign (Kirbus and Wendisch, 2024). PNSD data from all research flights of the BACSAM II campaign are available on PANGAEA (Simon et al., 2026). Sea ice data were retrieved from www.seaice.uni-bremen.de (Spreen et al., 2008). MODIS-TERRA satellite images were retrieved from https://wvs.earthdata.nasa.gov. This study has been conducted using E.U. Copernicus Marine Service Information; https://www.doi.org/10.48670/moi-00281.

Author contributions. DJS participated in the data collection and flight planning during the BACSAM II campaign, analyzed and interpreted the data, and wrote the manuscript. JS participated in the data collection and flight planning, discussed the data, and contributed to the writing. JH participated in the data collection and flight planning, and processed the position, nose boom, and humidity data. BK performed the trajectory calculations and supported their analysis. JM participated in the data collection, flight planning, and calculated the radiative heating/cooling rates. ZJ, MH, BW, LK,AMJ, and MW participated in the flight planning, and the data collection and related discussions. KW was responsible for the development of the MPSS system. AH and ZJ functioned as PIs during the campaign. AH, ZJ, SH, GCR, MW, and MLP contributed to the discussion of the results. FS conceived the study, functioned as PI during the campaign, and contributed to data interpretation and writing. All authors read and edited the manuscript.

Competing interests. The authors declare no competing interests.

Acknowledgements. We gratefully acknowledge the funding by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – Project Number 268020496 – TRR 172, within the framework of the Transregional Collaborative Research Center "ArctiC Amplification: Climate Relevant Atmospheric and SurfaCe Processes, and Feedback Mechanisms (AC)^3", as well as the support of the Alfred-Wegener-Institute, Helmholtz Centre for Polar and Marine Research through grant number AWI_PA_02172. The authors used a large language model (ChatGPT by OpenAI) solely to support coding tasks; all scientific reasoning, methodological decisions, analyses, and conclusions were developed independently by the authors.

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