# 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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## 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 $\approx$ 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 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

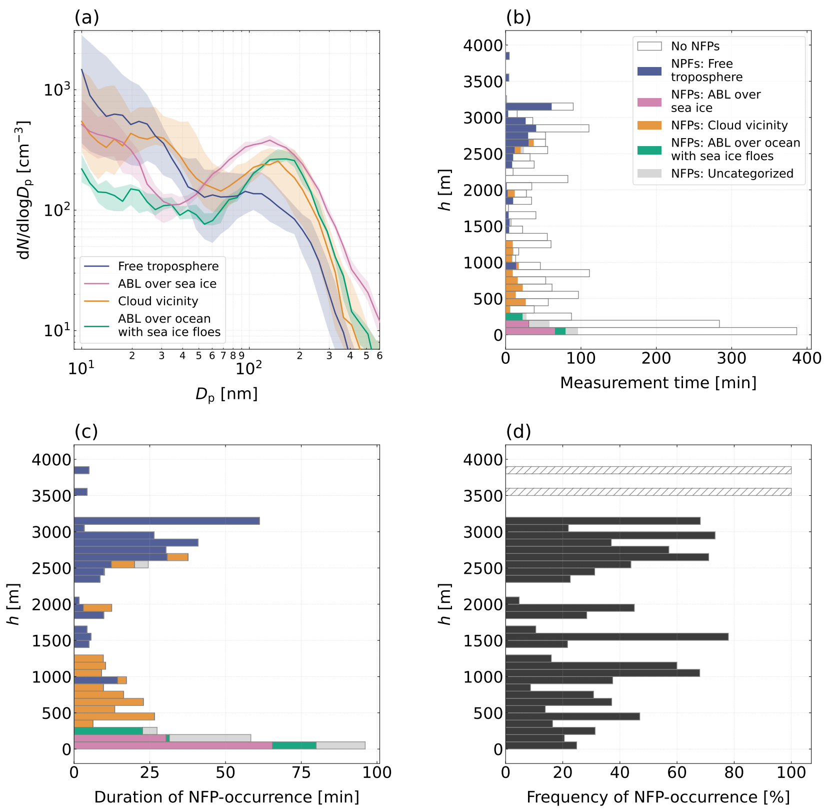


Figure R1: **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and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

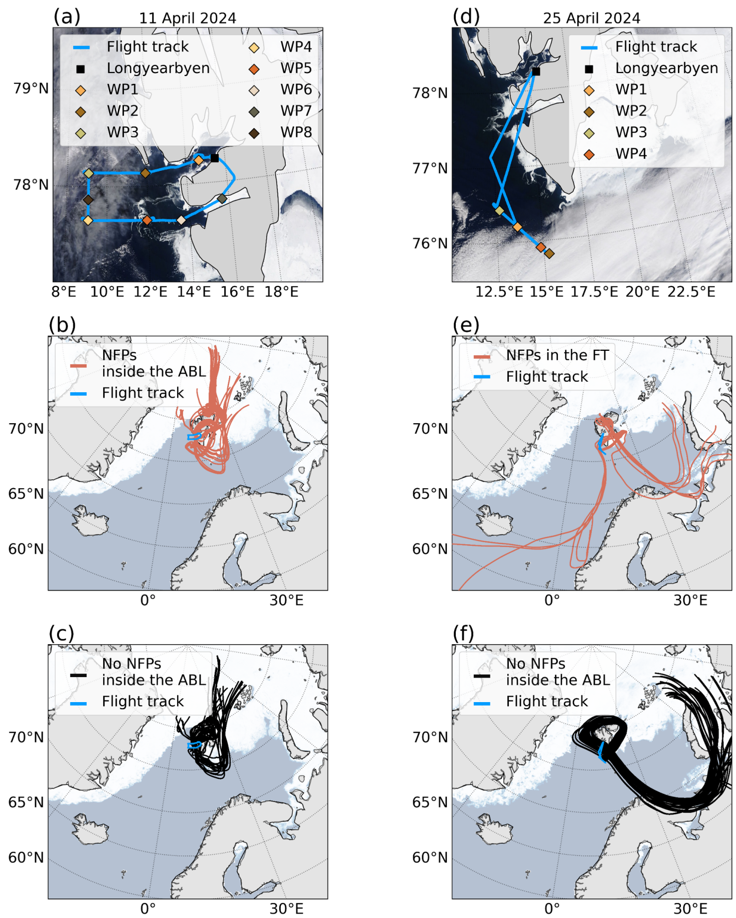


Figure R2: **(a)** Flight track of RF03 on 11 April 2024. **(b)** Five-day backward trajectories associated with newly formed particles (NFPs) inside the ABL on 11 April. **(c)** Five-day backward trajectories connected to PNSD measurements featuring no newly formed particles inside the ABL on 11 April. **(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 $\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. R3a, see Fig. R2a for the location of the WPs) with legs at $\approx$1300 m in the free troposphere (10:25 to 10:40 UTC) and inside the ABL at $\approx$240 m (10:50 to 11:05 UTC), $\approx$75 m (11:10 to 11:40 UTC), and again at $\approx$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 ($N_{10-20} \approx 25\,\text{cm}^{-3}$, $\approx 10\,\%$ 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. 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, $N_{10-20} \approx 50\,\text{cm}^{-3}$, 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

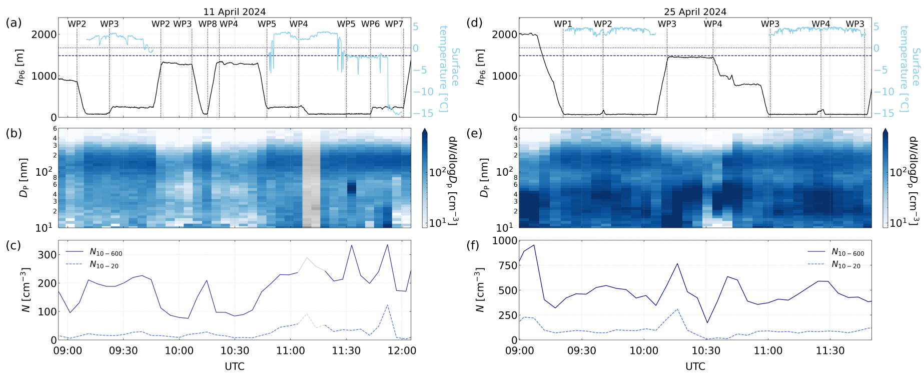


Figure R3: **(a)** P6 altitude during RF03 on 11 April 2024 (black line, left $y$-axis) as well as surface temperature (blue line, right $y$-axis). The dashed/dotted blue horizontal lines illustrate a surface temperature of 0°C and the freezing temperature of (salty) Arctic ocean water of −1.8°C. **(b)** 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 $\leq D_\mathrm{p} \leq$ 600 nm, solid line) and in the nucleation mode size range (10 nm $\leq D_\mathrm{p} \leq$ 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 $\leq D_\mathrm{p} \leq$ 600 nm, solid line) and in the nucleation mode size range (10 nm $\leq D_\mathrm{p} \leq$ 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 $\approx 67$ m were carried out inside the ABL over open ocean (9:20 to 10:05 UTC and 11:00 to 11:45 UTC, Fig. 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 $\approx 1400$ 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. 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

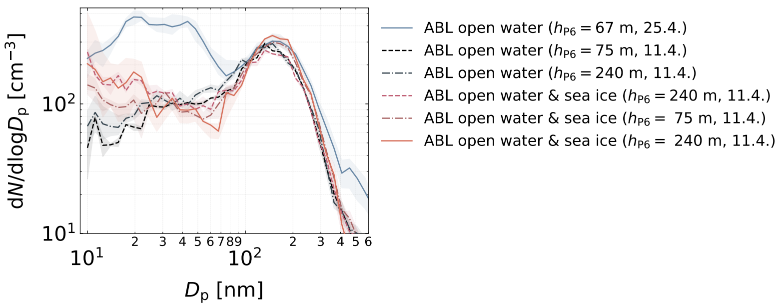


Figure R4: Median particle number size distributions during legs at constant altitude inside the ABL over completely ice-free open water (on 11 and 25 April, black and blue curves) and over ocean with both sea ice floes and open water areas present (on 11 April, red curves). The latter distributions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

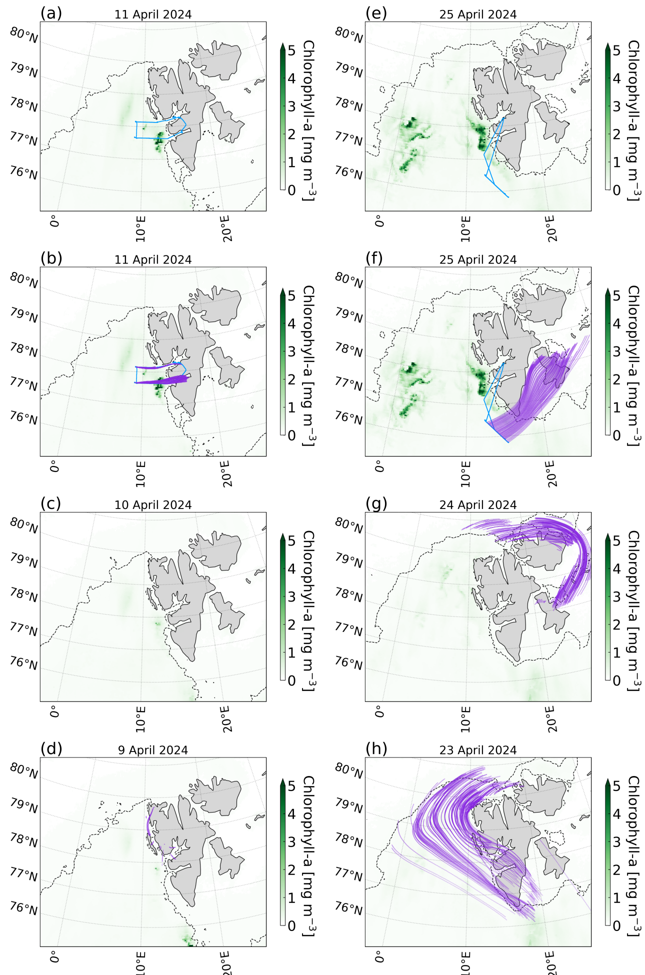


Figure R5: Flight track (blue), backward trajectories inside the ABL (violet), and chlorophyll-a mass concentrations (green) based on Copernicus-GlobColour data (Gohin et al. 2002; Hu et al. 2012) for the research flights featuring measurements inside the ABL over ocean on 11 and 25 April. The left column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

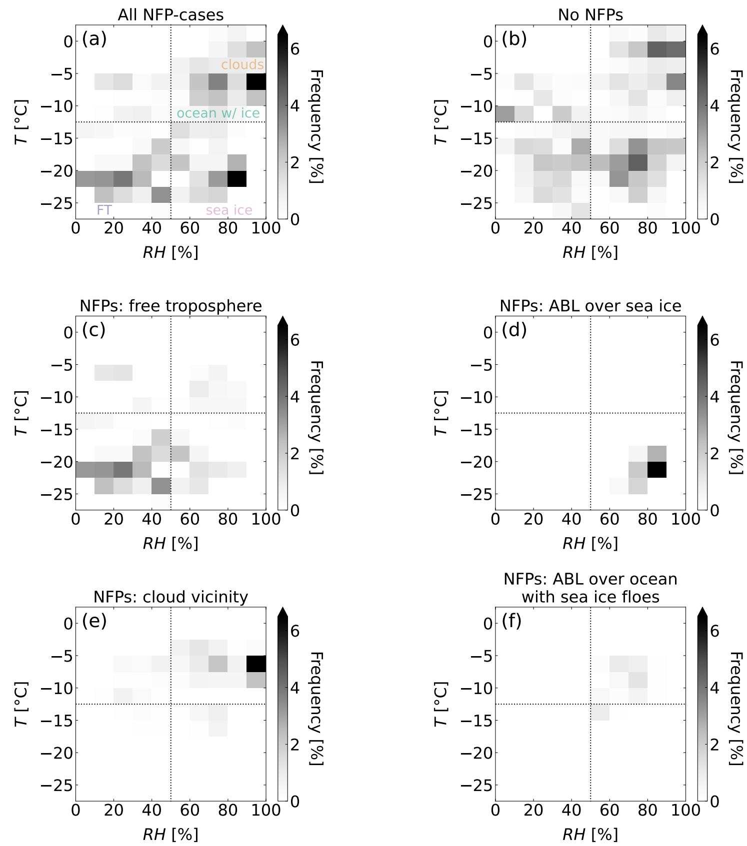


Figure R6: Temperature and relative humidity conditions during **(a)** newly formed particle-cases (NFP-cases) and **(b)** non-NFP-cases. The labels "FT", "sea ice", "clouds", and "ocean w/ ice" in panel (a) denote that within the corresponding quadrant, the measured temperature and relative humidity 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 $\sim 10$ 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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