Response to Editor and 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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Editor: Dear David Simon and co-authors,
I am pleased to inform you that, after addressing the minor revisions suggested by one reviewer (attached below), your article can be published in ACP. For the final decision, please submit an author's response and a revised manuscript based the points raised by the reviewer.
Best wishes, Martina Krämer

Author reply: We would like to thank you for handling the review process of our study. We have thoroughly revised the manuscript based on the remarks expressed by reviewer #1. Additionally, we have carefully checked the manuscript for typos.
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.

Reviewer #1: I want to thank the authors on their extensive work with the reviews, the manuscript has improved significantly. I have a few remaining points that are minor in execution, but crucial to the interpretation of the study, particularly my first comment.

Author reply: We thank the referee very much for reviewing our manuscript again. We have carefully revised the manuscript based on the listed points.

Please find our detailed response in the following paragraphs.

1. Following the review suggestion, there is now a more extensive description of the flight times and the atmospheric conditions, however, it is placed in the SI. The authors argue that relative frequencies of NPF cannot always be presented due to indistinguishable conditions, which I fully understand. However, I absolutely believe that the relative frequencies described in section S3 and Table 1 are an integral part of the data collection and must be presented in the main manuscript. There it is clear that more than double the flight time was spent in the FT compared to the other categories combined. This means that the numbers presented in lines 155-160 (e.g. 45% of NPF measured occured in the FT) in the main manuscript are misleading. They are true, but completely out if context. I understand that the boundary layer height cannot always be distinguish, but the authors should at the very least include the relative frequencies from Table 1, i.e., the relative NPF occurence in each category. If not, then also remove the frequencies as they do not mean anything standing alone.

We thank the reviewer for raising this point again. We agree that, without further context, the percentages mentioned in the original manuscript could lead to misinterpretation. We have revised the corresponding paragraphs in Sect. 3.1, where we now also include Table 1 (previously placed in the SI), which presents the total measurement times and relative frequencies of NFP-occurrence for the four environments. Please find the details in the revised version below (lines 155 to 189 in the revised manuscript):

[...] As mentioned earlier, newly formed particles were detected in a wide range of altitudes between approximately 60 m and 3900 m (Fig. R1b). Here, 60 m and 3900 m correspond to the minimum and maximum flight altitudes at which measurements were carried out (Fig. R1b). Consequently, we cannot rule out that newly formed particles could be present at altitudes even higher than 3900 m in the target regions during our campaign.

In the free troposphere, newly formed particles were detected at altitudes between about 900 m and 3900 m (Fig. R1b). These observations amount to measurement times of approximately 4 hours and 40 minutes, corresponding to distances flown of $\approx$ 1460 km. Inside the ABL over sea ice, at altitudes between roughly 60 m and 200 m, newly formed particles were identified during about 1 hour and 40 minutes of measurements ($\approx$ 350 km), and inside the ABL over ocean with sea ice floes, at altitudes between 60 m and 300 m, during approximately 40 minutes ($\approx$ 130 km). During about 2 hours and 40 minutes ($\approx$ 630 km) of measurements in the vicinity of clouds, newly formed particles were observed. Here, these particles were found at altitudes between 300 m and 2700 m (Fig. R1b).

Table 1 summarizes the durations of newly formed particle-occurrence for the four environments. Furthermore, based on ERA5 reanalysis data, we present 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 (see Sect. S3 for details). Additionally, relative frequencies of occurrence of newly

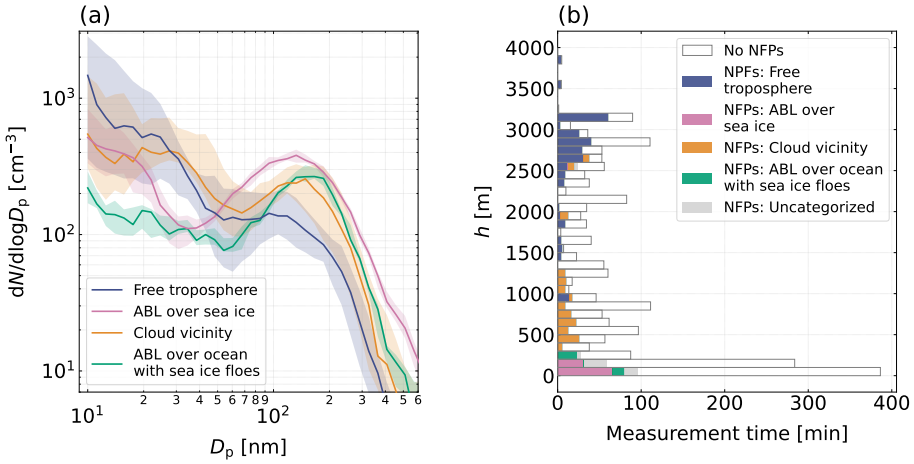


Figure R1: **(a)** Median PNSD of all measurements showing newly formed particles (NFPs) associated with one of the environments described in the main text: newly formed particles in the free troposphere (Sect. 3.2), inside the ABL over sea ice (Sect. 3.3), in the vicinity of clouds (Sect. 3.4), and inside the ABL over ocean with ice floes (Sect. S5). **(b)** Total measurement time per altitude (grouped into altitude bins with a width of 100 m). Colored bars illustrate the duration of newly formed particle-occurrence in each altitude bin, labeled by the different environments. White bars indicate the absence of newly formed particles. The data are shown as stacked bars.

formed particles for each environment are provided in Table 1. As discussed in Sect. S3, the ERA5-based values are associated with large uncertainties. Consequently, the total measurement times throughout the campaign as well as the frequencies of occurrence should be interpreted solely semi-quantitatively and only provide a very rough estimate.

It should be noted that, for all case studies described below, as well as for 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 considered 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 absence/presence of clouds close to the aircraft could not be verified, these observations are assigned to the unclassified cases (Sect. S3). Such observations represent 8 % of all PNSDs featuring newly formed particles (≈ 50 minutes, ≈ 200 km of measurements).

Naturally, during the campaign, sampling times and consequently numbers 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. R1b). For example, while measurements in the lowest 100 m were carried out for a total duration of almost 400 min (over a distance of about 1480 km), measurements at 3500-3600 m and 3800-3900 m were merely 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; Fig. R1b). 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. They rather provide a rough overview and represent (semi-quantitative) statistics of the observations made during the BACSAM II campaign.

Nevertheless, despite these limitations with respect to the statistical analysis, our measurements clearly demonstrate that newly formed particles were frequently found throughout the lower Arctic troposphere, and suggest that NPF in the Arctic occurred across large geographical areas and over a wide range of altitudes during our campaign in April 2024. [...].

Accordingly, we have slightly modified Sect. S3 to minimize redundancies. Please find the revised Sect. S3 below (lines 32 to 65 in the SI):

Due to the lack of, e.g., drop sonde and cloud microphysical measurements, data concerning the atmospheric stratification and the occurrence of clouds along the flight tracks are not continuously available. In other words, we cannot always unambiguously distinguish the airplane's 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, 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

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
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
Total measurement time [1] (based on ERA5)	25 h 10 min	8 h 10 min	3 h 30 min	1 h	2 h 10 min
Frequency of NFP-occurrence [1]	19 %	29 %	48 %	67 %	($>$ 100 %) [2]

1: Note that the ERA5-based values (i.e., total measurement time and, consequently, frequency of NFP-occurrence) should be considered semi-quantitatively only (see the discussion in the main text and Sect. S3 for details).
2: 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.

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 ($CWC = CLWC + CIWC$) 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. 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.

The values reported above are associated with rather large uncertainties as they are are not well suited to describe the prevailing conditions at the aircraft's exact position. 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 the classification of measurement periods featuring newly formed particles to the categories "free troposphere", "cloud vicinity", "ABL over sea ice", and "ABL over ocean with sea ice floes", including all case studies, we only considered measurements for which the prevailing conditions could be unequivocally determined based on the in situ data. If newly formed particles were detected but the conditions could not unambiguously be identified (e.g., because the ABL height could not be determined for the target region, or because the absence/presence of clouds close to the aircraft could not be verified by studying in situ video and radiation data), these observations are assigned to the unclassified cases.

2. My second point is regarding Figure 2d. The way the relative frequencies are presented here does not seem super useful to me, as there are many bins with very little measurement time that end up with large spikes in NPF frequency (most notably at 1500m). What do we learn from this figure? I would

suggest maybe binning the data differently, so that there is equal flight time in each bin and maybe move it to the SI.

Thank you for this comment! We have removed Fig. 2d from the manuscript entirely. Furthermore, we have removed Fig. 2c as well, as the data shown there was a zoomed in version of Fig. 2b and is therefore already shown anyways in Fig. 2b. Please find the revised Fig. 2 above in our response to the reviewer's comment #1 (Fig. R1).

3. Lastly, I think the response to review comment 15 regarding biological production should be reflected in a change in the text. It is still stated among the main conclusions that no indication of NPF over ice-free ocean was found, but without context.

Thank you very much for this suggestion! We agree and have included the following sentences in the concluding section (lines 465 to 468):
Furthermore, our analysis suggests that during the measurements carried out inside the ABL over open, completely ice-free ocean, no phytoplankton bloom occurred in the target regions, i.e., no indications pointing toward significant emissions of biologically-produced precursors were found. This might explain why no newly formed particles over open ocean were observed during our campaign.