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"

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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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