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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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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