Referee comments 1

Review of "Ice Nucleating Particle Concentrations over the Eurasian-Arctic seas" by Li et al., submitted to ACPD (egusphere-2025-2798)

This study presents a comprehensive dataset of ice-nucleating particle (INP) concentrations measured during the Arctic Century Expedition (August–September 2021) across the Barents, Kara, and Laptev Seas. The authors combine online (HINC) and offline (DRINCZ) immersion freezing measurements with aerosol chemical and physical analyses to examine INP abundance, sources, and variability in the Eurasian Arctic. The work expands observational coverage in a remote and data-scarce region and provides insights into the local versus long-range sources of Arctic INPs.

The manuscript is well structured and clearly written, with a logical flow from methods to results and interpretation. The inclusion of a focused case study contrasting low and high INP periods adds significant value by integrating aerosol measurements, chemical tracers, meteorology, and air mass origin to support the broader conclusions. This is a well-executed and much-needed observational study in a remote and understudied Arctic region. I recommend publication after minor revisions to address the points below.

We thank referee 1 for the nice summary and positive feedback on our manuscript egusphere-2025-2798. In response to the questions and suggestions, please find our answers and revisions listed below. **Referee comments are reproduced in bold** and author responses in normal font; *extracts from the original manuscript are presented in red italic* and *extracts from the revised manuscript in blue italic*.

1. **The use of 2-day back trajectories in Figure 7 provides useful context for identifying short-range influences but may be insufficient to fully assess the role of long-range transport, particularly for mineral dust or aged aerosols that can persist in the atmosphere for several days. While the authors conclude that local sources dominate INP variability, this conclusion may partly reflect the limited temporal scope of the trajectory analysis. Extending the trajectory duration could potentially reveal additional source regions or transport pathways not captured in the current analysis, such as high-latitude terrestrial dust or recirculated Arctic aerosols. As it stands, the statement of limited long-range influence, though plausible, remains somewhat constrained by the methodology. A brief discussion of this limitation and consideration of longer trajectory timescales would be welcome.**
   We thank the reviewer for this constructive comment. Indeed, the use of 2-day back trajectories focus on identifying local to regional influences on INP concentrations, particularly in the Arctic boundary layer, as trajectories were initialized near sea level. This approach was chosen to directly link near-surface measurements with recent surface interactions. We agree that this approach may not fully capture potential long-range transport events, such as those associated with aged mineral dust or recirculated aerosols that have residence times exceeding two days.

   To address this limitation, we have extended our back trajectory analysis to 7 days (see Fig. E3 for all measurements at -34 °C and -15 °C; and Fig. E4 for selected high and low N_{INP} cases at -20 °C, respectively, in the Appendix) and complement our discussion in Section 3.5 (see lines 335-338 in the revised manuscript): *"This finding is corroborated by extended 7-day back trajectories (see Fig. E3 in the Appendix). Notably, elevated N_{INP} observed near Novaya Zemlya at both -34 °C and -15 °C coincide with air masses characterized by prolonged residence over the western Siberian coast - a hotspot acting as a potent source for both mineral dust and biological particles as discussed previously."* and section 3.6 (see line 368 in the revised manuscript): *"…from the west Siberian coast, suggesting INP influence from both local source and long-range transport."*

2. **The weak correlations between INP and chemical tracers in Figure 4 limit the strength of source attribution to mineral dust or marine biogenic components. While the observed trends, such as modest associations with AlSiCa or sulfur, are directionally consistent with expected sources, the low**

correlation coefficients and lack of statistical significance in many cases suggest that the chemical composition data alone are insufficient to differentiate dominant INP types. This highlights the need for either more specific tracers (e.g., molecular markers) or multivariate approaches to better resolve complex and potentially co-varying source contributions.

We thank the reviewer for this critical comment. We agree that the correlation analysis between INP concentrations and bulk elemental composition (e.g., AlSiCa for mineral dust, P and S for marine biogenic sources) shows generally weak to moderate relationships and often lacks statistical significance at several temperatures. This indeed limits the strength of any direct attribution of INPs to specific sources based on the current data alone. The reviewer's point touches on two fundamental challenges of in-situ field measurements that are worth elaborating on.

First, as the reviewer correctly points out, the Arctic system is characterized by a mixture of multiple aerosol sources, many of which are highly dynamic in space and over time. Given the limited temporal and spatial coverage of observational data in the remote Eurasian Arctic, particularly over oceanic regions, our study was designed to serve as a first-order assessment using widely accepted tracer classes. However, we acknowledge that these tracers (e.g., bulk Al, Si, Ca, S) lack the chemical specificity required to isolate IN-active subfractions such as certain organics, biological macromolecules, or specific mineral phases, since our classification does not resolve the composition of individual particles or identify specific organic molecules known to be IN-active. Highly effective INPs may constitute only a tiny fraction of the total aerosol mass, and their specific chemical signatures can be diluted in a bulk sample, leading to weak correlations.

Second, field-based studies usually face unique challenges in disentangling source contributions due to complex atmospheric mixing, limited sample volumes, and operational constraints (e.g., storage and processing of filter/impinger samples). In a dynamic environment like Eurasian Arctic Ocean measured from a moving ship, air masses are rarely influenced by a single, isolated source. Instead, they contain a complex mixture of aerosols from marine, terrestrial, and biogenic origins that are often co-emitted or mixed during transport, making it statistically challenging to attribute a change in N_{INP} to any single tracer compared to a fixed-site measurement where a lot more data for a given location is available. The resulting weak or insignificant correlations reflect this complex, mixed nature of the aerosol population, where no single component likely dominates the INP population across all conditions. To more informatively reflect the intricate multivariate associations, both a larger number of independent samples and a broader suite of tracers (e.g., molecular-level organics, isotopic signatures) would be beneficial, which are, however, not available from our dataset from this ship-based expedition. Even with such data, multivariate source apportionment would remain challenging due to the relatively low INP concentrations and high variability of natural INP sources, particularly in Arctic environments.

To address this important point, we have now expanded the discussion in Section 3.3 to better frame the limitations of source attribution in our study and to highlight specific future directions that could enhance the quality of INP source identification in the Arctic, or generally, field studies (see lines 282-292 in the revised manuscript): *While directional trends between N_{INP} and elemental tracers (e.g., AlSiCa, P, and S) are broadly consistent with potential mineral dust and marine biogenic influences, the generally weak to moderate correlations and limited statistical significance at several temperatures suggest that bulk elemental composition alone cannot fully resolve the diversity of INP sources. This is due to the chemical and physical complexity of the ambient aerosol population that are influenced by multiple potentially correlated factors in the Eurasian Arctic, where marine, terrestrial, and biogenic particles frequently co-emit and mix during transport. Furthermore, highly effective INPs constitute only a small fraction of total aerosol mass and are not adequately represented by bulk tracer signals. To disentangle the overlapping terrestrial and marine contributions to INPs, particularly in the remote and dynamic Arctic oceanic environment, future field studies would benefit from the use of more specific molecular tracers (e.g., molecular organics, biochemical tracers like DNA and lipids, and isotopic ratios, etc.), single-particle analyses, larger sample sizes, and multivariate data approaches, the latter of which are not possible with the limited dataset we have from this single cruise.*

3. **The use of coarse elemental proxies (e.g., AlSiCa) without complementary biological markers (e.g., DNA, lipids) constrains the ability to resolve source types (marine vs terrestrial biogenic).**
While bulk elemental proxies offer broad insight into mineral and mixed biogenic inputs, we agree that they are not sufficient to resolve finer distinctions between marine and terrestrial biogenic INP sources. We acknowledge that incorporating more specific biological markers, such as DNA, lipids, proteins, or other molecular-level indicators, would provide a stronger basis for distinguishing between biogenic INPs of terrestrial and marine origin. However, such biological analyses were not available from the current study. Instead, we had additional heat treatment of the aerosol samples to attribute the heat labile (proxy for biological molecules) in another accompanying study (Li et al., 2025, doi: 10.1039/D4FD00160E).

4. **The discrepancy between impinger and PM10 filter data is well noted in Appendix A, but two important methodological issues remain unaddressed. First, the vertical offset between sampling inlets (2nd vs 6th deck) could introduce a height-related bias, particularly under stratified conditions where aerosol concentrations may vary significantly with altitude. Second, the potential for contamination from the ship's own exhaust emissions is not discussed, despite the low ambient aerosol background and proximity of the sampling locations to possible emission sources.**
We thank the reviewer for raising these two important methodological questions. We agree that these details are crucial for interpreting our results and will add them to the manuscript. We will address each point separately.

   1) We acknowledge that in a strongly stratified atmosphere, this vertical separation could potentially introduce a bias in the measured aerosol and INPs. However, the marine boundary layer is often well-mixed, which would minimize small altitude-related concentration differences for the aerosols sampled. In addition, the LVS showed higher INP concentrations than the impinger measurement which was at a lower altitude. Nevertheless, this is a methodological detail that needs to be mentioned. We now add a statement to Section 2.2.1 acknowledging the different sampling heights and the small potential for a related bias (see lines 86-91 in the revised manuscript): *The impinger and LVS were located on the 2nd and 6th decks, respectively, with a vertical separation of over 10 meters. Under stratified atmospheric conditions, especially near the ocean surface, the vertical variability of aerosol properties between different sampling heights cannot be entirely ruled out. However, given that both are in the boundary layer, we do not expect large differences in aerosol number and composition between the two sampling locations, except those introduced by the sampling method (impinger samples particles > 0.5 μm aerodynamic diameter).*
   2) Concerning the potential for contamination from ship exhaust emissions, we took active steps to minimize its influence. For the PM_{10} filter samples (LVS on the 6th deck), sampling was automatically halted whenever the wind direction was from the ship's funnel to avoid contamination. For the impinger and HINC measurements (on the 2nd deck), we used CPC data to identify time periods as being affected by exhaust plumes based on sharp transient spikes in total particle concentration, which were filtered out and excluded from INP analysis. These measures substantially reduce the risk of exhaust contamination in the reported data, although we acknowledge that low-level or short-lived influences may not be fully eliminated. A corresponding clarification has been added to Section 2.2.1 in the revised manuscript (see lines 91-96): *In addition, to minimize contamination from ship exhaust, specific procedures were followed for the two sampling locations. On the 6th deck, the PM_{10} filter sampling (LVS) was automatically paused whenever wind direction sensors indicated air flow from the ship's funnel. For impinger and HINC measurements on the 2nd deck, spikes in total particle number concentration measured by a CPC were used to identify exhaust plumes; these periods were flagged and removed from the dataset. These measures substantially reduce the influence of exhaust emissions on the reported INP data, although minor residual contamination cannot be fully excluded.*

5. **The weak to moderate correlations between INP concentrations and aerosol surface area shown in Figure 3 suggest that existing parameterizations may not fully capture the complexity of INP behavior in this environment. It would be helpful if the authors could expand the discussion to explore possible**

contributing factors, such as the potential diluting effect of non-IN-active sea salt particles or the presence of mixed aerosol types. A brief reflection on these mechanisms would strengthen the interpretation.
Excellent point. As the reviewer points out, sea salt particles, which are abundant over open ocean and IN-inactive at temperatures we measured, can contribute substantially to total aerosol surface area and thus dilute any direct relationship with INP concentrations. Moreover, the coexistence of multiple aerosol types (e.g., sea salt, biogenic organics, mineral dust) can lead to variability in IN-activity even at similar surface area concentrations, especially when effective INPs make up only a minor fraction of the total aerosol population.

To address this, we have expanded the discussion in Section 3.2 to reflect these factors and to clarify that aerosol surface area alone may not be a sufficient predictor of INP variability in mixed-source marine environments (see lines 262-265 in the revised manuscript): *Additionally, the high abundance of sea salt particles, which contribute significantly to total surface area but are largely inactive as INPs, could further dilute the relationship between N_{INP} and surface area concentrations. Moreover, the presence of mixed aerosol types with varying ice-nucleating efficiencies can further obscure any direct scaling with bulk surface area.*

Minor comments:

1. **Table 1, please clarify that CPC measures particle number concentrations.**
The entry of Function in Table 1 is changed to *"Particle total number concentration".*

2. **In Figure 2, the yellow circles, grey open triangles, blue bars, and pink diamonds all correspond to data at the same temperature points, but this is not immediately clear to the reader. Adding a guiding arrow or visual connector between the temperature axis and the symbol legend would help direct the eye and improve interpretability.**
We agree, the data points are only plotted at -15 °C; as such, they are only relevant for that temperature as plotted. To better navigate the reader, we add the temperature in the legend for the said data points to better guide the reader in the revised manuscript.

Reference

Li, G., A. Welti, A. Rocchi, G. Pérez Fogwill, M. Dall'Osto and Z. A. Kanji. Terrestrial and Marine sources of ice nucleating particles in the Eurasian Arctic (2025), Faraday Discussions, 258, 94-119, DOI: 10.1039/D4FD00160E