We sincerely thank the Associate Editor for overseeing the review process and both reviewers for their careful reading of the manuscript and their detailed, constructive comments. We have revised the manuscript accordingly and address each comment below, indicating in blue the changes made to the text where relevant.

1 Reviewer 1

1. *"It is not clear why various techniques are used to probe the INP efficiency".*

   The use of multiple techniques was motivated by complementary and distinct scientific needs that a single instrument could not fulfill. To clarify this, we have added an introductory paragraph at the beginning of Section 2.2 summarizing the contributions of each instrument:

   ...(geometric) diameter (D). To this end, the INA of the samples was investigated using four complementary techniques, each fulfilling a distinct and necessary function that could not be covered by a single instrument. AIDA (Section 2.2.1) and INSEKT (Section 2.2.2) were used in combination to extend the temperature range of the INAS density measurements. While AIDA operates at colder temperatures, INSEKT reaches warmer temperatures through offline filter-based freezing assays, together providing a more complete characterization of the temperature dependence of n_s. AIDAm (Appendix A) was used to characterize the temporal evolution of the INAS density at a higher temporal frequency than AIDA, by performing multiple consecutive expansions on aerosol sampled directly from AIDA. This is particularly relevant for the extended size range considered in this study, as larger particles are subject to gravitational settling inside the chamber, which could progressively alter the size distribution of the aerosol during an experiment and therefore affect the measured INAS density. The AIDAm time series therefore allowed the investigation of whether the INA changed with changing PSD. Finally, the IR-DROFA (Section 2.3) provided an independent cross-check of the INAS density using a bulk suspension approach combined with BET-derived specific surface area measurements. This approach is complementary to the aerosol-based surface area estimates used in AIDA and INSEKT, and allows us to compare the derived n_s values obtained with the different surface area estimation methods. The following sections describe each instrument and its specific measurement approach in detail.

2. *"Section 2.1: What was the detection efficiency of Malvern instrument. Did this instrument measured all the particles that went through the laser beam. Did the PSD have any errors/uncertainties?"*

   We thank the referee for this important methodological question. The Malvern Mastersizer 2000 is an ensemble laser diffraction instrument, which measures the collective diffraction pattern produced by all particles simultaneously present in the laser beam and does not measure particles via single-particle detection. Therefore, the concept of "detection efficiency" is not directly applicable in the same way as for optical particle counters. The key quality control parameter for this instrument is the obscuration (the fraction of laser

light blocked by the sample), which we maintained within the recommended range of 1–20% throughout all measurements. Furthermore, because the measurement principle is ensemble-based, all particles present in the illuminated volume contribute simultaneously to the recorded scattering pattern. The uncertainty in the PSD arises primarily from (i) assumptions made in the inversion of the Mie scattering signal (e.g., refractive index, sphericity of particles), and (ii) sample preparation. The Mastersizer 2000 typically achieves a repeatability of better than ± 1–2% in the D50 (volume median diameter) under stable conditions. Additional uncertainties may increase for irregularly shaped particles such as mineral dust. Nevertheless, Malvern PSDs were not used in any calculation presented in this paper. The main goal of showing these measurements was to contrast qualitatively the PSD measured by the Malvern right after sample segregation and the PSD obtained after injection of the sample into the AIDA chamber. To reflect this, we have extended the description of our Malvern measurements in the last paragraph of Section 2.1:

... subsequently, the particle size distribution (PSD) was measured using a Malvern Mastersizer 2000 (Malvern Instruments Ltd), which employs the laser diffraction method based on Mie scattering theory (ISO 13320-1). The instrument determines PSDs from the angular distribution of scattered light of an ensemble of particles rather than detecting individual particles; thus, all particles within the laser beam contribute to the measurement signal. The nominal measurable size range is approximately 0.02 μm to 2,000 μm. Ethanol was used as dispersant. The refractive index for the dispersant and particles was 1.360 and 1.520, respectively. The technique yields volume-based particle size distributions, reporting diameters of equivalent spheres assuming a spherical shape. Uncertainties in the retrieved PSD arise from assumptions in the inversion algorithm (e.g., refractive index), particle shape effects, and sample dispersion. Under controlled conditions, the reproducibility of laser diffraction measurements is typically within a few percent (ISO 13320), but variability may be higher for irregular particles such as mineral dust. Furthermore, reproducibility was further assessed through n=2 to 6 replicates per sample. The mean bin-wise coefficient of variation of the surface area distribution, defined according to $\overline{CV} = \frac{1}{N}\sum_i \sigma_i/\mu_i$, where μ_i and σ_i are the across-replicate mean and standard deviation of the surface area distribution, respectively, and N is the number of replicated measurements, was below 6% for 5 of 7 samples, reaching 12% and 6% for the Morocco samples (Figure 3b and c, respectively). This metric captures point-by-point shape variability across the full distribution. These results indicate high repeatability of the Malvern measurements for the analyzed samples. It should be noted that the PSDs derived from Malvern measurements were used to qualitatively compare the PSD obtained from freshly segregated samples with the PSD measured during the MICOS campaign inside the AIDA chamber, rather than being used in any calculation.

3. *"Abstract: We know that size plays an important role in ice nucleation and this study supports this premise. The abstract section could be improved, discussing why size plays an important role. It is also important to discuss why Moroccan sample showed not size dependency, but Icelandic showed subtle size dependence".*
We agree with the referee on this observation. The revised abstract now includes an ex-

plicit discussion of why larger dust particles are relevant for ice nucleation, as well as the mineralogical reasons behind the contrasting size dependence observed for the two sources, namely the constant K-feldspar content in Moroccan samples and the size-varying pyroxene fraction in Icelandic samples. The revised abstract reads:

Ice nucleation activity (INA) in the mixed-phase cloud (MPC) regime has been extensively studied. Nevertheless, most research has focused on particles smaller than a few micrometers, leaving the INA dependence on mineralogical composition and size poorly characterized for particles larger than ten micrometers in diameter. This gap is important because: (1) large mineral dust particles (LMDPs) can undergo long-range atmospheric transport and reach altitudes where MPCs form; (2) their mineralogical composition may differ from that of smaller particles, affecting the density of ice-active sites; and (3) larger particles are more susceptible to atmospheric aging through coatings by chemical and organic compounds, which can further modify their INA. Here, we used natural soil dust samples collected during field campaigns in Morocco and Iceland, covering particle sizes from fine to super-coarse dust, and characterized in terms of mineralogical composition and size. The samples' INA was measured in the AIDA and AIDAm cloud chambers and the INSEKT and the IR-DROFA freezing assays in more than 300 experiments. Moroccan samples exhibited INA comparable to that reported for K-feldspar in previous studies, with no dependence on particle size, consistent with the nearly constant K-feldspar fraction across all size bins. In contrast, Icelandic samples showed lower INA than in other studies using samples of similar composition, along with a subtle size dependence linked to the size-varying pyroxene content, which decreased with increasing particle size. Our findings elucidate the role of LMDPs in immersion freezing, and their relationship with mineralogy and size for low- and high-latitude dust sources.

4. *"Large particles often are observed coated with secondary compounds like organics and sulfates because they provide larger surface area. Are these coated particles poor INP? To further improve the implications of this study what are the limitations to using larger size particles? Some discussion along this line in the Introduction section would be useful to understand the atmospheric implications of large size particles"*

   We thank the referee for this pertinent observation. We note that this point was already partially addressed in the conclusions section, where we stated: "...For instance, larger particles might interact more strongly with chemical and organic components, potentially lowering or enhancing their INA." Nevertheless, we agree that expanding this discussion in the introduction would help to understand the role of larger particles in the atmosphere, and we have therefore added the following discussion to the introduction after line 51: Nevertheless, the atmospheric implications of large dust particles as INP remain subject to important uncertainties. Despite evidence for long-range transport, coarse and super-coarse particles on average have shorter atmospheric lifetimes than fine dust due to gravitational settling making their number concentration lower than those of fine particles (Adebiyi et al., 2023). Consequently, their contribution to bulk INP concentrations depends strongly on transport distance, removal rates, and their particle ice nucleation efficiency. Apart from the smaller number concentration, the large surface area of super-micron particles is prone

to coating with secondary compounds such as organics and sulfates. The effects of these coatings on particles' INA efficiency depend on the nucleation mode and coating material. In immersion freezing mode, coatings can suppress, leave unchanged, or enhance dust INA depending on their composition: studies found that secondary organic aerosol coatings had no significant effect on INAS density (Kanji et al., 2019), sulfuric acid coatings reduced INA with the strongest effect on feldspar (Augustin-Bauditz et al., 2014), and mineral dust internally mixed with biological material exhibited enhanced INA at warmer temperatures than pure dust (Augustin-Bauditz et al., 2016). These findings demonstrate that aerosol coatings can substantially modify the INA of mineral dust depending on the chemical nature of the coating material. Since these interactions occur at the particle surface, coating effects may become increasingly important for larger particles due to their greater available surface area and potentially more diverse distributions of ice-active sites.

2 Reviewer 2

1. *"I feel the restructuring of the paper and highlights the main points would improve the readability of the paper. Maybe move some of the more detailed derivations, intermediate equations, or sensitivity analyses to the Appendix and keep the main text focused on key steps and conclusions."*

   We thank the referee for this thoughtful suggestion and for taking the time to consider the overall structure of the manuscript. After carefully examining the manuscript structure we have decided that we would like to keep the current structure, and we would like to explain our reasoning. We consider central to the scientific discussion of the paper the derivation of the differential and cumulative INAS density (Section 2.7) and the analysis of the influence of nonlinear terms on n_s (Section 3.1). Specifically, the nonlinearity analysis in Section 3.1 is directly motivated by the extended size range considered in this study, which spans three orders of magnitude and makes the commonly used linear approximation potentially inadequate. We consider that keeping it in the main text ensures that the reader knows the conditions under which the standard approximation holds and where caution is needed. Furthermore, the mathematical framework established in Section 2.7 is repeatedly referenced throughout Sections 3 and 4, particularly in the discussion of the effective surface area approach and the size dependence of INA for Icelandic samples. Nevertheless, we acknowledge the referee's concern about readability. However, for the reasons outlined above, we are inclined to maintain the current structure, and we believe that the additions made in response to the other comments in this review further strengthen the clarity and scientific focus of the manuscript.

2. *"For Morocco, you state that INA shows no dependence on particle size, while for Iceland you find a "subtle" size dependence linked to pyroxene content. Given the central role of size in the paper, I suggest bit more quantitative discussion of this if possible. For example, include more explicit statistics for the size effect."*

   To provide a more quantitative assessment of the size dependence, we added "Section 4.3.5 Quantitative assessment of size dependence":

   We have computed pairwise comparisons of the ice active fraction between experiments sharing the same dust source but differing in particle size distribution. For each pair, we report the median $\log_{10}$ ratio ($\tilde{r}$) of frozen fractions at matched temperatures (temperature difference smaller than 0.7 K), defined as $\tilde{r} = \text{median}\left(\log_{10}\frac{FF_a}{FF_b}\right)$, where FF_a and FF_b are the ice active frozen fractions at matched temperatures for experiments a and b, respectively, as reported in Figures 13 and 14 for Morocco and Iceland, its interquartile range ($\text{IQR}_{\log}$), the geometric mean ratio ($G = 10^{\tilde{r}}$), and the two-sided Wilcoxon signed-rank test p-value. The Wilcoxon signed-rank test is a non-parametric test of the null hypothesis that the median log ratio is zero, i.e. that the two experiments are statistically equivalent. A p-value below 0.05 indicates that the null hypothesis is rejected at the 5% significance level, meaning that a difference as large as observed would occur by chance in fewer than 5% of cases if the two experiments were truly equivalent.

For Morocco (Table 1), the four pairwise comparisons yield geometric mean ratios ranging from 0.664 (experiments 27 and 26) to 2.138 (experiments 28 and 29), indicating that at the typical matched temperature, experiment 28 has 2.138 times higher frozen fraction than experiment 29. Only one pair reaches statistical significance (experiments 28 and 29, p=0.016). The remaining three pairs show no significant difference (p=0.062–0.375), and the median log ratios are similar to their IQR_{log}, e.g. experiments 5 and 16 have a $\tilde{r}$=0.277 and IQR_{log}=0.180. This shows that the ice active frozen fraction shows no systematic dependence on particle size for Moroccan dust: the differences between size fractions are modest, statistically weak, and do not point consistently in any direction.

For Iceland (Table 2), all five pairs with sufficiently large number of matched points (n$\geq$ 6) yield statistically significant differences (p=0.008–0.031), with geometric mean ratios ranging from 2.1 to 8.3. The pairs with the largest G values correspond to experiments 31 and 34 and experiments 11 and 3, with geometric mean ratios of 8.296 and 5.043, respectively. As shown in Figure 14, experiments 11 and 31 contain a larger contribution from particles with diameters smaller than 0.8 μm to the total surface area than experiments 3 and 34. In contrast, Figure 14c shows that experiments 17 and 22 have nearly identical surface area fractions, which is reflected in their statistics: although experiment 17 is consistently above experiment 22 (p=0.008), the ratio varies considerably across the temperature range (IQR_{log}=0.306, $\tilde{r}$=0.360), with a geometric mean ratio closer to those observed for Moroccan paired experiments. It is worth noting that for all other Icelandic pairs the IQR_{log} is at least half the value of $\tilde{r}$, indicating a more consistent and strong offset across temperatures. Furthermore, the case of experiments 31 and 33 is noteworthy: despite experiment 33 having a larger surface area contribution from particles smaller than 8 μm than experiment 31, experiment 33 exhibits systematically lower ice active frozen fractions at comparable temperatures, a difference that is statistically significant (p=0.016, $\tilde{r}$=0.477, IQR_{log}=0.237). It is not clear why this pair deviates from the general trend, whereby a larger contribution from smaller particle diameters to the total surface area is associated with higher ice-active frozen fractions at equivalent temperatures.

The one pair that does not reach significance (experiments 30 and 6, p=0.125) is constrained by n=4 matched points, for which p=0.125 is the minimum achievable p-value by construction of the exact Wilcoxon test, regardless of size effect. However, the geometric mean ratio of 5.399 and IQR_{log} of 0.208 for this pair are consistent with the other Iceland pairs. Taken together, the Iceland comparisons provide quantitative evidence for a size-dependent ice nucleation activity signal that is both statistically significant and physically large.

Table 1: Derived statistics for the pairwise logarithmic ratio of ice active frozen fraction between experiment number a ($\exp_a$) and experiment number b ($\exp_b$), defined as $\log_{10}(FF_a(T)) - \log_{10}(FF_b(T))$, computed at matched temperatures (temperature difference smaller than 0.7 K) for the Morocco experiments shown in Figure 13. n is the number of matched ice active frozen fraction pairs used in the calculations, $\tilde{r}$ is the median logarithmic ratio, $\mathrm{IQR}_{\log}$ is the interquartile range of the log ratio ($Q_{75} - Q_{25}$), $G = 10^{\tilde{r}}$ is the geometric mean ratio, and W_p is the two-sided Wilcoxon signed-rank test p-value.

$\exp_a(\#)$	$\exp_b(\#)$	n	$\tilde{r}$	$\mathrm{IQR}_{\log}$	G	W_p
5	16	5	0.277	0.180	1.893	0.062
20	9	7	0.054	0.069	1.131	0.297
27	26	7	-0.178	0.152	0.664	0.375
28	29	7	0.330	0.220	2.138	0.016

Table 2: Derived statistics for the pairwise logarithmic ratio of ice active frozen fraction between experiment number a ($\exp_a$) and experiment number b ($\exp_b$), defined as $\log_{10}(FF_a(T)) - \log_{10}(FF_b(T))$, computed at matched temperatures (temperature difference smaller than 0.7 K) for the Iceland experiments shown in Figure 14. n is the number of matched ice active frozen fraction pairs used in the calculations, $\tilde{r}$ is the median logarithmic ratio, $\mathrm{IQR}_{\log}$ is the interquartile range of the log ratio ($Q_{75} - Q_{25}$), $G = 10^{\tilde{r}}$ is the geometric mean ratio, and W_p is the two-sided Wilcoxon signed-rank test p-value.

$\exp_a(\#)$	$\exp_b(\#)$	n	$\tilde{r}$	$\mathrm{IQR}_{\log}$	G	W_p
11	3	8	0.703	0.220	5.043	0.008
31	33	7	0.477	0.237	2.998	0.016
31	34	6	0.919	0.222	8.296	0.031
33	34	6	0.326	0.151	2.117	0.031
17	22	9	0.360	0.306	2.293	0.008
30	6	4	0.732	0.208	5.399	0.125

3. *"Moroccan samples showed no dependence on particle size, likely because the K-feldspar content remained constant across all sizes. How confident are you about your composition measurements for larger particles?"*

   This is an important point, which Section 4.1 partially addresses; however, following the referee's comment, we agree that a more thorough discussion was needed and have extended it as follows:

   ...Furthermore, externally mixed feldspar represented 3.2% of all particles by mass, and a 10th of it was estimated to correspond to K-feldspar. It is important to note that this is externally mixed K-feldspar and there is additional internally mixed K-feldspar, however, this cannot be isolated by the technique used in Panta et al. (2023). An important finding of the Moroccan samples analyzed was that the relative amount of externally mixed feldspar and K-feldspar was invariant across particle size. In contrast, the mass fractions of complex quartz-like and feldspar-like particles increased with particle size, ranging from 5% to 18% for feldspar-like particles. However, determination of the composition of larger

particles is inherently limited. While the elemental composition obtained is most probably representative for the whole particle, as all of the particle's cross-section is scanned, the SEM-EDX technique used in Panta et al. (2023) identifies only the dominant mineral type in each particle. No actual phase identification is carried out, all particle classes are therefore labeled "-like" to reflect similarity in elemental fingerprints rather than confirmed mineral phases. This ambiguity is particularly relevant for larger particles, as the proportion of internally mixed aggregates increases with particle size, making the attribution of the EDX signal to a specific mineral such as K-feldspar increasingly uncertain. Nevertheless, a specific particle size threshold beyond which classification becomes uncertain is not defined, and the limitation is described qualitatively as a progressive effect with increasing particle size.

4. *"Emphasizing what is new for high-latitude dust parameterizations (e.g., capturing pyroxene associated variability, broader spread in ns) and what that implies for Arctic/boreal mixed-phase cloud simulations."*

   Highlights about the new contributions of our study to low- and high-latitude sources were summarized in the section conclusions. Following the referee's comment this section is extended to explicitly address some of the most relevant points regarding the implication of our study:

   ...Additionally, the fit obtained with the effective surface area for Iceland agreed with previous measurements of similar origin or mineralogical compositions. Based on the above, we can conclude that the results presented here have direct implications for the parameterization of INA from HLD sources in atmospheric models. Existing parameterizations used in cloud microphysics schemes are predominantly derived from LLD and MLD sources. Our results show that applying these parameterizations to Icelandic dust would overestimate INP concentrations by up to 1.5-2 orders of magnitude (Figure 10b). This overestimation would translate into an excessive glaciation of Arctic and boreal mixed-pase clouds with potential effects on cloud lifetime, optical depth, and precipitation. Additionally, the observed pyroxene-associated variability with the INA underscores that a single deterministic parameterization may be insufficient for Icelandic or volcanic related sources. This variability is related to the heterogeneity in pyroxene content and surface properties, and suggests that representing HLD as a distinct aerosol class with source-specific and surface area dependent INP parameterizations in regional Arctic models would improve the fidelity of simulated mixed-phase cloud properties. To this end, the effective surface area approach introduced here - which scales the particle surface area to the size-dependent pyroxene fraction - offers a physically based framework to incorporate mineralogical variability into INP parameterizations for HLD sources.

5. *"You showed an important comparison of INAS densities from AIDA and INSEKT and show systematic offsets. However, it is sometimes unclear whether observed differences in INA are driven more by sample properties (size, mineralogy) or by methodological differences between instruments."*

   This important point is clarified in section 3.2 with the addition of:

...closer than the INSEKT data (slope -0.64, Figure 9d) to the parameterization of Sanchez-Marroquin et al. (2020) (slope ≈ -0.46). It is important to note that the comparisons in Section 4 were always performed between experiments using the same instrument, ensuring that methodological offsets between AIDA and INSEKT did not affect our conclusions regarding sample properties. Nevertheless, we acknowledge that the systematic offset between AIDA and INSEKT affects the absolute n_s values and therefore the parameterizations derived from the combined dataset (Table 4, Figure 10). Although its cause remains unclear, this offset is consistent with previously reported inter-instrument discrepancies (Hiranuma et al., 2015) and introduces an additional source of uncertainty in the proposed parameterizations (as accounted for in the confidence intervals). Disentangling instrument-related from sample-related contributions to the observed spread in the overlapping temperature range has been the subject of previous studies (Hiranuma et al., 2015) and would require dedicated intercomparison experiments using identical aerosol samples measured with both techniques, which is beyond the scope of the present study. For this reason, the fits obtained with each individual technique are also presented (Figure 8 and 9).

6. *"In its current form, the manuscript focuses heavily on instrument intercomparisons and systematic biases, ns calculations, giving it the tone of a technical report rather than a science-driven study. It would strengthen the paper to expand the scientific discussion, particularly regarding the implications of these biases."*

   We thank the referee for this constructive input. We understand how the inclusion of several technical sections could give this impression. For this reason, we would like to clarify the reasoning behind their inclusion and highlight the scientific content of the manuscript. The technical sections, such as section 3.1 which is devoted to the influence of nonlinear terms on n_s, and the AIDAm analysis were included because the extended size range considered here (which is one of the main novelties of the study) required methodological checks that needed to be documented. However, we believe that the paper contains substantial scientific discussion that goes beyond the instrument intercomparison. For example, sections 4.1 through 4.4 provide a detailed mineralogy-driven interpretation of the INA results, including the role of K-feldspar for Morocco, the pyroxene content and its size dependence for Iceland, and the novel effective surface area concept. Furthermore, the conclusions are science-driven: no dependence of INA on particles size was observed for Morocco, while a size dependence is present for Icelandic samples, and the effective surface area approach produces physically meaningful results. Nevertheless, we acknowledge that the scientific implications of our results, for example their relevance for atmospheric modeling, or the role of inter-instruments offsets, could be discussed more explicitly. To this end, the additions made in response to the other comments in this manuscript strengthen and clarify these points.

7. *"The link to atmospheric relevance part could be improved. For example, how using your Morocco vs Iceland parameterizations would change predicted INP concentrations in a simple idealized case?"*

   We note that the parameterizations presented in Table 4 for Morocco and Iceland allow us

to compare the predicted INP concentrations. This simple idealized case is added in the conclusion section:

To illustrate the atmospheric relevance of our parameterizations, we present a simple idealized comparison of predicted INP concentrations using the Morocco and Iceland parameterizations from Table 4, grounded in atmospheric observations of Icelandic dust from Sanchez-Marroquin et al. (2020). In that study, airborne Icelandic dust surface area concentrations during dust events were reported. Using a representative dust surface area concentration of 50 μm^2 cm^{-3} (Table S1 in Sanchez-Marroquin et al. 2020), the Morocco parameterization predicts INP concentrations approximately 3.5 times higher than the Iceland parameterization at 255 K, increasing to approximately 46 times (1.7 orders of magnitude) at 247 K. This demonstrates that the choice of parameterization becomes increasingly important at colder temperatures relevant for mixed-phase cloud glaciation. However, it is important to emphasize the idealized and illustrative nature of this comparison. Implementing source-specific INP parameterizations in atmospheric models poses several important challenges. For example, Icelandic dust is an episodic source with high temporal variability (Groot Zwaaftink et al., 2017), requiring accurate representation of dust emission events in regional and global models. Additionally, n_s requires knowledge of the PSD evolution of the emitted dust, making it necessary that models independently track Icelandic dust. Furthermore, the relative contribution of Icelandic dust to the atmospheric INP population compared to other dust sources, and the altitudes and temperatures at which this contribution is most relevant, remains to be quantified through dedicated modeling studies. We plan to address these challenges in future work combining the parameterizations derived here with a dust transport model and source-specific tags for high-latitude sources.

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