We thank the reviewers for the time they put in to provide comments and suggestions that ultimately improve the scientific rigour of this paper. Responses can be found below, and any changes to the manuscript and supplementary information are noted.

Reviewer 1 - Comments

1. The main questions concern the experimental methodology to calculate the active site density as a function of T, ns(T), and in particular the role of the size distribution (SD). For a given type of aerosol, the characteristics of the SD, reported in the supplementary information, show some variability from one experiment to another. Can the authors comment on this situation? Did they have control over the variation in particle size and particle concentration, or do these parameters vary more or less randomly?

   Response: *For a given material, the total aerosol concentration was intentionally varied between experiments to extend the range of particle surface area concentrations and thereby access a broader range of ns(T) values. While these changes influenced the total particle number concentration, the overall shape of the distributions remained broadly consistent for each sample material. A new subsection (SI Section S3.1) has been added to the Supplementary Information describing the experimental variability in aerosol concentration and size distributions to provide clarity on this matter. This new subsection also contains information relevant to Reviewer 1, comment 2.*

2. Could these variations in SD between experiments be the cause of the significant variations sometimes observed in the active site density measurements? Indeed, the nucleation process depends on particle size, yet this parameter is no longer taken into account in the calculation of ns(T), as this is expressed in terms of the total surface concentration of particles, which renders the size parameter irrelevant. Could the authors shed some light on this aspect?

   Response: *In calculating ns(T), we assume that ice-nucleating activity is proportional to particle surface area and that ns(T) is not dependent on particle size. Although it is possible that particle composition, and therefore ice-nucleating activity, may vary with particle size, evidence for a dependence of ns(T) on particle size is lacking in the literature. However, to minimise potential biases, we aimed to maintain a similar shape of particle size distributions between experiments for a given sample material but make changes to the overall concentration. Because the wash-off and drop-on filters were sampled simultaneously from the same aerosol population during each experiment, any variability in particle size distribution affected both techniques equally and*

therefore does not influence the study's primary comparison. This clarification has now been added to SI Section S3.1 in the same section addressing reviewer 1 comment 1.

3. Section 2 Methods (page 4 to page 12) : In section 2.1, there is a subsection "2.1.1 Filter sampling" (line 163) Is it correct ? I suggest renumbering subsection '2.1.1' as section 2.2 and renumbering the following sections as 2.3, 2.4 ...

   Corrected

4. Line 199, page 7 and line 211, page 8 : If I am correct, units of k(T) should be cm-3 °C-1 and not cm-3 °C.

   Corrected

5. Line 226 : "ns(T) for each individual experiment plotted with its uncertainty can be found in SI Section S2". - From my point of view, this sentence should be placed at the end of this section after introducing this ns(T) parameter.

   Corrected

6. Can you specify how the uncertainties on ns(T) (drop-on and wash-off techniques) are calculated as shown in the SI section? Do the uncertainties come from the µL-NIPI method?

   Response: *We agree that additional clarification was warranted regarding the uncertainty associated with ns(T). The following statement has been added following Eq. 7: The uncertainty associated with the background-subtracted k(T) values was propagated through the calculation of $K(T)$ and subsequently through to N_INP(T) for both the drop-on and wash-off techniques. The uncertainty on ns(T) was calculated by combining the N_INP(T) uncertainty with that of the total particle surface area, s, using standard error propagation in quadrature.*

7. Line 230: Eq(6) page 9, I suggest to specify also the unit of ns(T) , cm-2, even if it is evident and it can also be found in Figure S3 of the Supplementary Information.

   Corrected

8. Line 298 : "To establish". The sentence seems to be incomplete.

   Removed

9. Line 321, Table 1. I propose listing the samples in the rows of Table1 in the order in which they appear in the main text: Dry Snomax, Copper River Dust, Agricultural.

   Corrected

10. Line 412, figure 6(a) : I suggest resizing the horizontal axis from -20 to -10°C in order to distinguish more clearly between the data relating to drop-on and wash-off techniques.

    Corrected

Reviewer 2 - Comments:

1. For the wet samples, a medical nebuliser was used and after evaporation, dry aerosol particles remained in the chamber. However, it is not completely clear how the size distribution, concentration and composition of these particles were controlled during the experiments. Were the aerosol PSD and number concentration actively controlled, or only measured afterwards with APS/SMPS and used for normalisation? It would be useful to show or discuss the temporal stability of the aerosol during filter sampling and PINE measurements, including possible changes due to nebuliser output, reservoir depletion, wall losses or particle settling.

   Response: *The aerosol number concentration was intentionally varied between experiments to obtain a wider range of aerosol surface area concentrations and corresponding ns(T) values. During individual experiments, the aerosol generation methods were selected to provide a relatively stable particle source. There were gradual decreases in aerosol concentration due to the depletion of the nebuliser reservoir. In some experiments, these decreases were partially mitigated by repositioning the nebuliser. We have added SI section S3.1 and Figure S3, which provide additional information with a time series of the aerosol surface area concentration over the experiments.*

2. For the dry Snomax experiments, the authors mention that electrostatic charge may have affected the size distributions. From a metrological point of view, it

would be good to discuss if using an aerosol neutraliser, for example Kr-85 or X-ray, before filter sampling or PINE measurements could improve reproducibility in future experiments.

Response: *We agree that an aerosol neutraliser may help limit the effect of static on sample materials such as the dry Snomax and would be helpful in future experiments. We add the following to section 4.1: "The use of an aerosol neutraliser prior to sampling could potentially reduce variability associated with particle charging and improve the reproducibility of the generated aerosol in future experiments."*

3. The use of active site density, ns(T), is appropriate, but it depends directly on the aerosol surface area concentration. Therefore, the APS/SMPS merging and the assumptions used to convert aerodynamic and mobility diameters into volume-equivalent diameters are very important. The authors should better explain the choice of density and dynamic shape factor. In the Supplement, some values seem to be selected to obtain consistency between APS and SMPS data, but the effect of this choice is not really quantified. In particular, assuming $\chi = 1$ for all filtrates may be a simplification. A short sensitivity analysis showing how ns(T) would change for $\chi = 1.1$ or 1.2 would make the results more robust.

Response: *For the wet-dispersed samples, the density and dynamic shape factor were estimated because literature values are not available for these samples. A dynamic shape factor of χ=1 was assumed for all wet-dispersed samples because aerosol particles generated by nebulisation originate from initially spherical droplets and were therefore expected to remain approximately spherical following evaporation. The filtrates contained material smaller than 0.2 um, making direct estimation of particle density challenging. Hence, values were chosen that produced a reasonable fit between the SMPS and APS data. We have added a sensitivity analysis to the supplementary information (new section S4.1) in which the APS/SMPS merging procedure was repeated using a reasonable range of shape factors and particle densities. The shape factor, χ, was increased by 20% while the density, ρ, was varied by +/- 20%. Although the reviewer's concern primarily relates to the wet-dispersed samples, the analysis was applied to all materials in this study. The results indicate that the calculated ns(T) values are more sensitive to the assumed shape factor than the density. However, for all sample types, the changes introduced by a change in χ and ρ were substantially smaller than the differences observed between the wash-off and drop-on techniques. Furthermore, because the wash-off and drop-on filters were collected simultaneously from the same aerosol population, both techniques used the same aerosol surface area estimate. This means*

uncertainties associated with APS/SMPS merging assumptions propagate equally to both techniques and do not affect the comparison between techniques. We therefore conclude that the differences reported between the wash-off and drop-on techniques are robust to reasonable uncertainties in particle shape factor and density.

4. Equation 7 also needs clarification. If the distribution is dS/dlogDp, then the total surface area should be integrated over logDp, not directly over Dp. Maybe the calculation was done correctly in logarithmic bins, but the equation as written is confusing.

   Response: *Equation 7 has been changed to integrate over logDp, as we did in the calculations.*

5. The comparison between wash-off and drop-on also includes different filter materials, different pore sizes, different droplet geometry and different sample processing. I suggest adding a short paragraph acknowledging these metrological limitations.

   Response: *While the wash-off and drop-on techniques differ in several aspects, such as material, pore size, sample processing, and droplet geometry, the purpose of this study was to evaluate the techniques as they are most commonly implemented in the literature. These methodological differences form part of the comparison between the two techniques. We believe that keeping the techniques consistent with previous literature was a strength of this study, rather than a limitation. The two techniques are described in detail in sections 2.2 and 2.3.*

6. In addition, the two filter samplers were placed at different positions in the chamber, which was mixed with a fan, but it is not clear if the two sampling positions were tested to be equivalent. Were the filter types alternated between positions, or were duplicate samplers with the same filter tested? If not, this should be mentioned as a possible limitation.

   Response: *We evaluated potential positional bias by testing the equivalence of the two filter sampling lines using agricultural soil dust and cellulose (which was ultimately not included in the study). PTFE filters were placed in both filter holders, and the resulting fraction frozen data showed no systematic differences attributable to sampling location (within approximately 1 °C). In addition, the filter type assigned to each filter holder was alternated throughout the*

experimental campaign to minimise any residual bias. We have added a description of these tests to Section S2 of the Supplementary Information.

7. The manuscript states that, when the background-subtracted k(T) falls within the background, the sample value is set to zero. I think this treatment may bias cumulative spectra, especially for samples close to the blank. It would be clearer to report detection limits, upper limits or confidence intervals instead of forcing these values to zero. The authors should also clarify how many blanks were used for each technique, whether they were distributed across different experimental days, and how the blank uncertainty was propagated into the cumulative INP spectra.

Response: *We acknowledge that setting background-subtracted values to zero may introduce a small low bias at the highest temperatures where sample activity approaches the background signal. However, because the nucleating activity tends to increase steeply with decreasing temperature, the influence on the overall freezing spectrum is minimal. The uncertainty associated with the background subtraction is still retained through the error propagation procedure and is reflected in the reported uncertainties in ns(T). It is not possible to systematically determine a detection limit (in ns(T)) since this depends on variables unique to each experiment. The method we use in this study is consistent with previous peer-reviewed studies (Barr et al., 2023; Raif et al., 2024).*

We have expanded SI Section S1 to clarify the frequency and treatment of handling blanks. Handling blanks were collected after the chamber was cleaned/flushed and before the introduction of sample aerosol. Three blank runs were collected for Copper River dust experiments and agricultural soil dust experiments, while one blank was taken for pollen filtrate, Snomax filtrate, lichen filtrate, agricultural soil filtrate, and sea surface microlayer experiments. No handling blank was collected before the dry Snomax experiments. The blank measurements were collected on different days throughout the experimental campaign following routine chamber cleaning. The mean blank spectrum was used for background subtraction, while the standard deviation of the blank spectra was used as the uncertainty associated with the background correction and propagated through the calculation of the cumulative INP spectra.

8. Page 12, Sec 2.4: The sentence “To establish ….” appears incomplete or incorrectly joined. Please revise.

   Removed

9. Line 219: "Eq 4 to calculate the INP concentration for the wash of technique" should be "wash-off".

   Corrected

10. Page 13, Sec 3.1.1: “contains derived from Pseudomonas syringae” I guess is meant “contains material derived from Pseudomonas syringae”.

    Corrected

11. Page 15, Sec 4.1: “dry-disperse samples” should be “dry-dispersed samples”.

    Corrected

12. Figure 6 caption: “activty” should be “activity”.

    Corrected

13. Figure 7 title and caption: “Sea Surace Microlayer” should be “Sea Surface Microlayer”.

    Corrected

14. Page 18, Sec 4.4: “particularly above 22 °C” perhaps is “above -22 °C”

    Corrected

15. Ensure consistency in temperature degree symbols (e.g., "-10 °C" vs "-10 ◦C" used in different sections).

    Corrected

16. Section 1: “Pue de Dôme” should be “Puy de Dôme”.

Corrected

17. line 245: Use “GmbH” rather than “Gmbh”.

Corrected

18. Use either “airflow” or “air flow” consistently.

Corrected

19. Use “litres per minute” or “liters per minute” consistently.

Corrected

20. Units in the supplementary tables show sampling volume as “L−1”. This should be “L” if the column is sampled volume.

Corrected

21. In the supplement, it is referred to experiments from “Table S2 to Table S8” but the dry Snomax table appears as Table S1. Is it left out for some reason?

Corrected