

First of all, I would like to thank the authors for their responses to my comments and the revisions made to the manuscript. The new version places greater emphasis on the methodology, which is satisfying regarding the purpose of this paper. This new version also includes better discussion on statistical analysis. However, in my opinion, the manuscript still requires improvements for potential publication. Specifically, it appears that (1) authors did not pay enough attention to details, (2) authors can further develop Sections 2.4, 2.5 and 2.6 that are the ones actually focusing on the novel coupling method and (3) authors must imperatively improve the discussion on statistical and error analysis of the technique started in Section 2.5.2 to provide readers a real understanding of the limitations of this method and potential improvements.

(1) Attention to detail, which was a major concern in my initial review, remains insufficient. For the figures, many of them require better formatting, clarity, and descriptions (e.g., many figures contain spectra that are misaligned, inconsistent in font size, or even cut off). Additionally, several figures include elements, text, or numerical values that are not properly explained in the captions. For the main text, several terms are utilized but not carefully defined. For the references, there are inconsistencies between the style of the listed publications. These issues must be addressed to improve the overall quality and readability of the manuscript.

(2) The coupling method, which should be the main focus of the manuscript as it represents the novelty in this work, requires further development. The authors state that they have placed greater emphasis on the methodological discussion while shortening the case study, making the manuscript more suitable for AMT. While these changes are noticeable, Sections 2.4, 2.5, and 2.6, which should form the core of the manuscript, must be expanded with a more detailed discussion and further explanation of the method. Additional figures would be beneficial in guiding the reader through the methodology.

(3) The statistical analysis and evaluation of uncertainties remain limited and require further discussion. Relying on ambient measurements (the case study) for statistical analysis is, in my opinion, an inadequate approach for rigorously assessing the method. Analysing aerosols generated under controlled laboratory conditions would have been preferable. In Section 2.5.2, the authors state that, for the case study, only an average of 30% (ranging from 13% to 50%) of identified INPs with FRIDGE were analysed using SEM. Among the remaining 70%, 45% were excluded due to excess aerosol loading on the wafer, while 25% were blank positions. For the 45% related to aerosol loading, high particle concentration increases the likelihood of multiple particles within the 50  $\mu\text{m}$  radius used for SEM analysis, making many positions non-analysable. However, a more in-depth discussion of this issue is needed, including an evaluation of different aerosol concentrations or sampling durations to identify possible improvements. Unfortunately, such an assessment is not feasible within the current case study framework, significantly limiting the statistical evaluation of the technique. Similarly, the issue of blank positions (25%) is not discussed at all, which is very concerning. This lack of discussion limits the transparency of the method's performance. Furthermore, the FRIDGE-SEM analysis cycle is not repeated, meaning no statistical validation through multiple similar measurements is provided.

### **Specific comments**

-L.221: "information on their chemistry". It is very vague, please refer to elemental composition, as it is only what EDX provides.

-Fig.1: I strongly recommend not mixing the style between subfigures, as A1, B1, B2 and C1 are schematics while B3, C2 and C3 are experimental results. If results are added to a subfigure, I expect them to be explained in the text and legend. In B3, what does TA, TB and TC mean? What is the x-axis? In C2, what is class A, class B, etc.? Explain why in figure C1 why you used a straight line for BSE and SE while you used wavy line for EDX.

-L.163: “ice formation can regularly be observed at temperatures at or below -30°C.” I highly doubt that ice formation above -30°C is never observed. It would be very useful to include blank results in the supplementary information to support this statement.

-L.188: “it is important to keep the three laser-engraved crosses on the wafer surface visible during the FRIDGE measurement.” Which ones? Please refer to figure in SI.

-L.215: “with a minimum size of 30 pixels proven to be useful” elaborate, explain why it is useful. If you refer to Schrod et al. 2016, cite the publication.

-L.218: “as the center of the detected bright area” what do you mean? Explain better what is bright.

-L.225: “To reduce this uncertainty based on an imperfect radial symmetry, the ice crystal position calculation should be performed on the basis of FRIDGE images, that show the ice crystals in a state close to activation.” I don’t understand this part. Do you mean that you need to take images before ice crystal formation? Or what do you refer to as “close to activation”?

-L.228: “calculated for SEM” what do you mean by calculated? The sum of all ice crystals?

-Fig.2: What does  $y$ ,  $R^2$ , and  $p$  represent? These parameters must be explicitly explained, even if they seem obvious.

-L.245: “solid-state detector (SSD), providing the distribution of elements on the particle by backscattered electrons (BSE) giving information on homogeneous or heterogeneous distribution of elements and on inclusions.” why don’t you provide examples of such analysis?

-L.248: “and origin” what do you mean by origin?

-L.253: “As the internal SEM coordinate system is centered around the origin in the middle of the stage aligning the axes to the directions of mechanical movements, it is necessary to perform a coordinate transformation to link the SEM coordinates to the coordinates defined by the crosses in the previous step.” I don’t understand this sentence, please be more explicit and pedagogical.

-L.254: “Based on a calibration image, which indicates the marked center points from the previous ice crystal identification step.” Did you consider to add a Figure to illustrate the calibration process from FRIDGE to SEM, I find it difficult to follow properly all the steps. At least in the SI?

-L.266: “In this context, a radius of 50  $\mu\text{m}$  has proven to be useful.” I think this can be further discussed. You can develop on the changes seen with different radius and show the statistics. As it is now, the choice of such radius sounds very vague.

-L.279: “possible particle drift during the processing in FRIDGE.” Could you elaborate on this? Additionally, could particle drift also occur when transitioning from FRIDGE to SEM-EDX analysis? Is there different possibility of drift based on particle size?

-L.287, “even if the aerosol concentration is known, it is difficult to specify a suitable collection volume in advance, as the ratio of potential INPs to the total aerosol also plays a role.” In L.270–285, you mentioned that SEM analysis is only feasible if no other particle is present within the 50  $\mu\text{m}$  radius. This shows that the feasibility of the analysis is not a matter of how many INPs are on the filter but rather how spaced the particles are. Thus, I think information on mass loading or recommended sampling time can be provided by the authors. This information can also be accompanied with statistical analysis of occurrence of multiple particles within 50  $\mu\text{m}$  radius. Furthermore, in Schrod et al. 2016, some guidelines about sampling for FRIDGE measurement is provided. As an AMT paper, I am expecting some guidelines for applying the method proposed here.

-L.291: “identification rate” you introduce a new term; a clear definition is needed.

-L.293: “INP identification rate was calculated to be 30% (ranging from 13% to 50%).” Is this based on all measurements from case study? This is crucial information; it needs further discussion.

-L.293:” identified the presence of multiple particles at 45 % of the locations (ranging from 7 % to 81 %)” why such a high variation? Is there any correlation with sampling time or aerosol concentration? You need to discuss these values.

-L.294: “While the remaining 25% (ranging from 2% to 66%) were found to be blank positions.” This part also needs further clarification and discussion. How were these values determined? Does this come from particles drifting? If yes, why such a high variation?

-L.298:” In most cases, the small number of clearly identified INPs still allows general statements to be made, e.g., about the most frequently occurring characteristics of INPs” you need to discuss that further. Why would I believe you? What are most cases? Can you provide a lower limit?

-Fig. 3: The spectra require a y-axis and should be replotted, as the Si spectra appear to be cut off. The x-axis has inconsistent font sizes, making the figure unsuitable for publication in its current form. Also, I first thought the image resolution 20 x 20  $\mu\text{m}$  was the size of the grid on the FRIDGE image, please add some scale to avoid confusion.

-L.312: “determine the mixing state of a particle” In cases of mixing state, how are INPs classified? Is the composition of the main particle that is assumed to be the INP?

-L.312: “surface properties” what kind of properties?

-L.327: “chemical characterization” why chemical or not elemental composition?

-L.345: “Carbonates can contain, in addition to carbon and oxygen” why on Figure 6 the ratio of signal for carbon and oxygen (C:O) is not 1:3 for carbonate ( $\text{CO}_3$ )?

-L.378:” Sulfates are mainly characterized by the presence of sulfur and oxygen.” Similar question here, why is there not a ratio S:O of 1:4 in Figure 6?

-Fig.6: Adjust the axes, align the spectra, why does the spectra don't start from zero?

-Fig.7: The images must be numbered and explicitly referenced in the text.

-L.409: “across the three activation temperatures,” and RH.

-L.412:” so the error of the concentrations given here is also in this range.” Why not adding error bars on the Figure?

-L.417:” Their concentration varied between 0.1 and 1  $\text{stdL}^{-1}$  for most of the time” what is the collection volume if I compare it to the background value of FRIDGE 0.1 $\text{L}^{-1}$  for 100L volume sampled.

-Fig.8: The blue color is difficult to distinguish, especially when a red triangle is placed over it. Consider improving contrast for better readability. Also, in my opinion, this figure is not directly relevant to the main focus of this paper: the coupling method. This figure can easily be removed from the manuscript and be replaced by more attention to Sections 2.4, 2.5 and 2.6.

-L.429:” Overall, based on the parameters described in Sect. 2.5.2, we were able to clearly identify and characterize the associated INPs for 200 ice crystals.” You mentioned in the same section that the identification rate is 30%, so you were able to characterize 200 from 600 ice crystals, no?

-L.437:” INP chemistry” be more specific.

-L.454:” Mineral components” why not proving all spectra in the SI?

-L.469:” Carbonaceous particles” why not proving all spectra in the SI?

-L.479: "Other particle classes" why not providing all spectra in the SI?

-Fig.9: I recommend labeling "during the Saharan dust event" with the letter (c) for clarity.

-L.512: Explain the calculation of the projected area diameter.

-Fig.10: This figure needs uncertainties. Please compare the amount of analysed INP with SEM-EDX compared to total INP number detected with FRIDGE.

### **Technical corrections**

-L.204: "humidity settings" change to relative humidity.

-L.221: "FRDGE" please change to FRIDGE

-L.234: "These Positions" no capital letter.

-L. 242: "Environmental Scanning Electron Microscopy (ESEM)" no capital letters to keep consistent with other abbreviations in the manuscript.

-L.572: "which ich assigned to" is to ich

-L.406: "GAW" what does it stand for?

-L.582: "It has been shown, that this position calculation works reasonably well". Who showed that, you?

-L. 594: "ice-active particles" why not INP?