Author's response to Reviewer 1

First of all, we would like to thank the reviewer for reading our revised manuscript and commenting on some technical improvements.

In the following, the reviewer comments are written in bold and our answers in italics. Text passages from the revised manuscript are in quotation marks, modified or newly added passages are marked in green.

Suggested technical corrections

L46: Please remove the first parentheses and use "e.g.," instead

Changed as requested.

"In addition to the prevailing environmental conditions, i.e., temperature and humidity, the potential for an INP to be activated depends on individual particle properties, e.g., surface imperfections (Kiselev et al., 2016), chemical composition and specific chemical properties, crystal structure, coating (Kanji et al., 2008), etc., as well as its atmospheric processing, including potential agglomeration or pre-activation (Marcolli, 2017)."

L78: Change "IR" to "IRs"

Changed as requested.

"In this case, the particles are heated after separation so that the water evaporates and ice residuals (IRs) remain."

L96-100: Please reshape these sentences, which are confusing and might mislead the readers. One can understand that the false INPs are inevitable, however, based on the statement here, the number of false INPs seems to be several orders of magnitude more than real ones. This brings issues regarding data quality and reliability.

We have rewritten the sentences and adjusted the perhaps somewhat drastic statement they contain. The new section reads as follows:

"These methods typically analyze large numbers of INPs/IRs. However, the major challenge of all these methods is that due to the extremely low number of INPs within a sampled air volume compared to the much higher number of non-INP particles (ratio ~ $1/10^4$ – $1/10^6$), the separation must be carried out with a very high accuracy. Conclusions about the chemistry of INPs may not be entirely accurate, since there is no way to distinguish particles that have been falsely separated as INPs from real INPs afterwards. There is also the risk that additional artifacts can be introduced into the INP fraction during the multi-step process. This problem is partially illustrated in the comparison of the chemical analysis of the INPs/IRs fraction by three different methods in Worringen et al. (2015)."

L101: Change "INP/IR" to "INPs/IRs"

Changed as requested.

"This problem is partially illustrated in the comparison of the chemical analysis of the INPs/IRs fraction by three different methods in Worringen et al. (2015)."

L110: Add a statement, e.g., the coupling has not yet been described in detail in previous studies

Changed as requested.

"Details of the FRIDGE method were described by Schrod et al. (2016), but the coupling has not been described in detail in previous studies."

L122: Change "INP" to "INPs". Go through the manuscript and revise when necessary.

Changed as requested (see the following sentence). We also went through the whole manuscript and changed it if necessary.

"Based on the definition by Cziczo et al. (2017) we refer to the identified particles as INPs, as they were activated under defined conditions after the collection of the total aerosol and not sampled as ice crystals."

L125: I would still argue the particles measured by SEM are IRs rather than INPs. As pointed out in the following sentence, these particles have undergone changes in FRIDGE. Such change includes ice activation and at the end of each measurement cycle, the ice crystal will evaporate before the measurement by SEM, therefore the remaining core should be IRs.

We can understand the reviewer's point of view. The nomenclature for INPs and IRs always depends on the perspective and is a matter of definition. For the reasons given in the text, we have decided to stick with the chosen nomenclature, but have mentioned in the text that a different perspective is also possible.

"However, some of the INPs analyzed with SEM may have undergone changes due to the measurement procedure in FRIDGE, and thus be announced as IRs. But we assume that these changes are of minor importance for the main INP classes that we can analyze with this method (see Sect. 2.6), which is why we have decided to continue referring to them as INPs."

L222: Change "x" to multiplication symbol. Go through the manuscript and revise all.

Changed as requested.

Author's response to Reviewer 3

First of all, we would like to thank the reviewer for reading our revised manuscript. The feedback and detailed comments helped us a lot to expand the technical chapters of the paper and to describe the method in a more focused way. Through the comments, some formalities could be improved.

In the following, the reviewer comments are written in bold and our answers in italics. Text passages from the revised manuscript are in quotation marks, modified or newly added passages are marked in green.

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.

We revised many of the figures in terms of clarity (Fig. 1, Fig. 6, Fig. 10, Fig. 11, Fig 12 – numbers referring to the new manuscript) and formatting (Fig. 6, Fig.7, Fig. 9, Fig. 10, Fig. S3 – numbers referring to the new manuscript). We went through all figure captions, and revised them if appropriate. We tried to explain all the used elements and numerical values in the captions. We hope, that they are now clearer and better to understand.
We went through the whole manuscript, trying to identify terms which are not properly explained. If this was the case, we added a definition.
Details for individual changes can be found in the specific comments below and in the manuscript with track changes.

All the references were revised with a citation software and the format given by the Copernicus website. In order to maintain the clarity of the document, these changes have not been marked in the track changes.

(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.

The method sections have been improved and extended as suggested. The identification process (Sect. 2.4) is now described in more detail regarding the coordinate systems. A figure has been added (Fig. 3a – number referring to the new manuscript) to visualize the INP identification in the coordinate systems.
Also, the coordinate calibration to recover the positions in SEM (Sect. 2.5.1) is now explained more understandable, in more detail and illustrated by a new figure (Fig. 3b-d number referring to the new manuscript).
A new chapter (Sect. 2.5.2 – number referring to the new manuscript) containing a numerical simulation of the effect of different parameters (total particle number, activated fraction, position uncertainty) on our INP identification process was added. This model study should give the reader a more detailed insight in the sensitivity of the method to different parameters and limitations. Also, it can help to derive recommendations for an optimal application of the method.
We also improved the discussion on ambiguous and blank positions as well as on the identification rates (Sect. 2.5.3 – number referring to the new manuscript).
For a detailed description of the individual points, please refer to the following specific comments.

(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 µm 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.

For the statistical analysis of the method and the influence of different parameters (total particle numbers on the wafer, activated fraction and position uncertainty) on our identification process, we added a model study (Sect. 2.5.2 – number referring to the new manuscript), as the case study is not perfectly suited for such an evaluation as you already noted. The discussion on multiple particle and blank positions has been extended (Sect. 2.5.3 – number referring to the new manuscript).

About the repetition of the coupling experiment, we would like to say the following:
The individual steps of the coupling procedure (FRIDGE, identification of ice crystal center points, recovery in SEM) were subjected to replication experiments. Schrod et al. (2016) showed that the average variation of the ice crystal number in FRIDGE is about 20%. The identification of ice crystal origins and coordinate calculation by our counting algorithm was also repeated. However, as this is an automated process, it always provides the same output with the same input, in this case the same coordinates.
The position of the coordinates in the SEM depends essentially on the calibration points. In the manuscript we give a value of 20 µm for the inaccuracy due to the limited resolution of the FRIDGE images. Repeated calibration on the basis of the calibration picture resulted in a deviation of ± 10 µm. As this deviation does not exceed the inaccuracy due to image quality, it was not considered further in the manuscript. In general, each new calibration of the wafer results in a slight change in the position of the calibration points; in random tests, the particles were found in a slightly different position to the identified coordinate.
To repeat the whole experiment procedure is not that easy, as it is not simply transferring the wafers between the two instruments. The oil from the FRIDGE experiment has to be removed, as it would otherwise affect the EDX analysis in SEM. This process is carried out manually using ethanol and special laboratory wipes, but is, however, a source of contamination. While possible lint on the wafer can be clearly identified as artifacts in the SEM, they can cause a bias in the following FRIDGE activation cycle. Another problem may be caused by the SEM/EDX analysis. Although SEM/EDX is generally regarded as a non-destructive method, ice nucleation properties react quite sensitively to the surface properties, which is why these can be altered by the introduction of electrons.
As a consequence, from our point of view, there is less potential for errors and it is more representative to repeat the individual steps than the whole multiple step procedure.

With regard to potential laboratory experiments we would like to state the following:
We can understand the reviewer's point of view, that laboratory experiments may be a good chance to evaluate a method. For the coupling, our aim was to develop and evaluate the method as closely as possible to field conditions. As a typical problem with ice nucleation measurements is that the devices work well under laboratory conditions but have problems in the field under real conditions. In many cases, field and laboratory measurements are also not directly comparable. The FRIDGE chamber itself has been subjected to many evaluation experiments over the years.

Specific comments

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

Changed as requested.

"The activated INPs can subsequently be characterized by SEM to gain information on their elemental composition, morphology and size (Fig. 1C)."

-Fig.1: I strongly recommend not mixing the style between subfigures, as A1, B1, B2 and C1 are schematics while and 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.

A1, B1 and C1 are schematic drawings of the three instrumental steps of the procedure (sampling, FRIDGE and SEM). In order to give the reader a meaningful overview of our method, B2 and C2 show exemplary results of a real analysis. B3 and C3 should be used to give the reader an idea how the overall result of the method looks like. These graphs are NOT based on real data and are therefore also considered to be schematic, which is why the legend is also schematic. TA, TB and TC are placeholders for different activation temperatures, similarly Class A, Class B etc. in C3 are placeholders for different INP classes. The date is shown on the x-axis in B3.
In the SEM, the SE and BSE detectors map the electron contrast by secondary/backscattered electrons (straight lines), while the EDX detector registers electromagnetic radiation (wavy line).
However, as a result of this discussion, Fig. 1 has been simplified. Only the schematic drawings of the aerosol collection, ice activation in FRIDGE and the analysis in SEM are now included. The presentation of the results has been verbalized for the sake of clarity.

-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.

It was already mentioned that there are three laser-engraved crosses on the wafer surface in Sect. 2.1., where the substrate is described. For clarity we have added a reference to Fig. 1 and the FRIDGE picture in the supplement (Fig S1).

„For coupling the INP activation experiment to the single particle analysis by SEM, it is important to keep the three laser-engraved crosses on the wafer surface visible during the FRIDGE measurement (see Fig.1 and Fig. S1 from the supplement). "

-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.

In fact, our ice crystal identification algorithm uses the basic parameters of the FRIDGE INP counting software, which are given in Schrod et al. (2016). As we modified this section more extensively, the thresholds are given now a bit later in the text (see next comment). There, we added also the reference to Schrod et al. (2016).

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

Bright area refers to the accumulation of pixels that exceed a previously defined brightness threshold and are therefore recognized by the software as ice crystals. We have adapted the wording accordingly.

"The software identifies the image with the highest number of ice crystals for each measurement cycle and tags the ice crystal positions in the images as the center of an area exceeding a size threshold of 30 pixels with a brightness threshold of 30 of 256 on a scale stretching from the darkest to the brightest recorded signal (Schrod et al. 2016). If two adjacent ice crystals are separated by at least one pixel that falls below the brightness threshold, ImageJ detects two separate areas and determines a center point for each of these areas. If this is not the case, a center point is determined for the entire area."

-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"?

In our case, "close to activation" means that ice growth has already begun, but the crystals have not yet grown large. We have adapted the wording accordingly.

"To reduce this uncertainty based on an imperfect radial symmetry, the ice crystal position identification should be performed on the basis of FRIDGE images showing the ice crystals in a state shortly after the initial activation."

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

The wording was misleading in this case. It refers to the positions that the software has "calculated", i.e., identified. We have changed the wording slightly to make it clearer.

"Figure 2 shows a comparison between the number of ice crystals counted by FRIDGE (parameters from Schrod et al. (2016)) and the number of ice crystal positions identified for the SEM analysis by the image analysis procedure described above."

-Fig.2: What does y, R2, and p represent? These parameters must be explicitly explained, even if they seem obvious.

We added explanations to the capture.

"Figure 2: Comparison of ice crystal numbers counted by FRIDGE with the number of identified positions for SEM analysis by the image analysis software. The linear regression is represented by the black line, the 1:1-line is shown in red. The regression equation relates the number of positions identified for SEM (y) to the ice crystal numbers counted by FRIDGE (x). It is given along with its coefficient of determination (R^2) and its p-value."

-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?

We provide examples for heterogeneous element distributions later in Fig. 6 and Fig 7(numbers referring to the new manuscript). At this point we only want to describe shortly the basics of SEM and some relevant capabilities of the technique.

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

Here origin was meant in terms of source. We changed the wording accordingly.

"The EDX provides an elemental composition of an individual particle, which can be used to attribute the analyzed particles to different classes of compositions and sources."

-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.

The whole section was reformulated. We tried to be more precise in the explanation using a new figure illustrating the different coordinate systems (for more details on the figure, see the next comment).

"To find the identified position for each ice crystal / INP by SEM, its normalized coordinates [X_n/Y_n] (Fig. 3a) from the ice crystal identification step (Sect. 2.4) must be converted to instrument specific coordinates [X_{ESEM}/Y_{ESEM}]. This is necessary because the internal SEM coordinate system is a Cartesian coordinate system centered on the middle of the stage, with the x and y axes aligned to the directions of mechanical movements (Fig. 3c). Based on a calibration image (Fig. 3b), showing the defined calibration points from the previous step (Sect. 2.4), these calibration points are obtained in the SEM coordinate system. Based on the internal SEM coordinates for the calibration points, all INP coordinates [X_n/Y_n] can be converted to internal SEM coordinates [X_{ESEM}/Y_{ESEM}]. It is highly important to locate these calibration points with the highest possible precision, since the position of each ice crystal in the subsequent analysis is calculated based on this calibration. Manual calibration provides

the highest precision, as the systems use different physical imaging processes yielding largely different image resolutions and contrast mechanisms. The high magnification of the electron microscope (Fig. 3d) in conjunction with the surface sensitivity of electron emission, in contrast to the limited resolution of the FRIDGE calibration image (Fig. 3b) produced by visible light reflection, impair any precise automated calibration. Due to the limited resolution of the FRIDGE images of about 20 × 20 µm, the calibration leads to a positioning uncertainty of the same scale."

-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?

We added a new figure (Fig 3 – number referring to the new manuscript) showing the coordinate system for the ice crystal identification step (Fig 3a), a calibration image (Fig. 3b), the Wafer on the SEM stage with the cartesian SEM coordinate system (Fig. 3c) and how a calibration crosse looks like in the SEM (Fig. 3d). We hope, that this figure helps the reader to understand the coordinate calibration step.

-L.266: "In this context, a radius of 50 µm 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.

The dependence of the search radius on various parameters such as positioning accuracy, number of particles on the wafer and INP fraction was investigated in a model study (Sect. 2.5.2 - number referring to the new manuscript). The results show a clear variability of the optimal radius, especially with variation of the positioning accuracy and the total substrate loading.
Besides the discussion in the new modeling section, the sentence in Sect. 2.5.3 (new manuscript) was also revised.

"Based on the simulations and empirical values, a radius of 50 µm around the identified coordinate was chosen as a standard search radius for the following INP identification."

-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?

(We decided to change the notation from particle drift to particle shift to avoid confusion with the drift problem sometimes encountered in SEM, which has other physical reasons.)

Particle shift in FRIDGE means a possible displacement of the particle due to ice growth or sublimation of the ice after a measurement. Such shifts are generally possible and cannot be excluded, but their extent and influence on our method can be reduced by various arrangements, e.g., by limiting ice crystal growth and avoiding the liquid phase in FRIDGE (mentioned in Sect. 2.3). Furthermore, if the identification of the coordinates for the ice crystal centers is based on the last measurement series from FRIDGE, multiple displacement due to repeated ice growth is excluded. Whether a potential shift is size dependent has not yet been investigated, and might be difficult due to the unknown original position. We have included a section on the influence of potential particle shift on the results in the discussion of identification rates (Sect. 2.5.3 – number referring to the new manuscript).

"In the case of particle shift being the reason for the blank position, there is a potential for a bias in particle frequencies or size distribution, if the potential for a particle to shift would correlate with particle class or size."

Of course, a particle shift can also occur during the transfer of samples from the FRIDGE to the SEM and cannot be completely ruled out. We do everything possible to avoid this (avoiding tilting, shaking, etc.).

-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 µm 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 µm 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.

While it is true that the total amount of particles on the wafer is crucial for the unambiguous assignment of individual INPs in the SEM (this can be also seen from the model), the absolute number of INPs also plays a role, e.g., for counting statistics at the lower end or ice-coverage at the upper. As the absolute number of INPs on the wafer depends on the proportion of INPs in the total aerosol, which is typically unknown prior to sampling, a compromise must be found between an amount of aerosol that allows for the unambiguous identification of as many particles as possible and a sufficient number of INPs to be deposited on the wafer. The whole section on discussing sampling parameters has been revised (including guidelines for applying the method) and now reads as follows.

"The total number of INPs that can be unambiguously attributed to an ice crystal origin is significantly influenced by the total wafer loading (INPs and non-INPs), which is determined by the sampling parameters (e.g., flow rate, sampling time, deposition efficiency) in combination with the aerosol concentration and aerosol properties. Even if the aerosol concentration is known, it is difficult to specify a suitable collection volume in advance due to the priori unknown fraction of INPs. Besides the total number of particles on the wafer, which is decisive for the chance to identify one particle in the defined radius and influences significantly on the ratio of falsely identified INPs (see modelling data in the supplement – Fig. S2), the ratio of potential INPs is also important to ensure that enough INPs are deposited on the wafer for suitable counting statistics. Based on the modeling, we would consider a value of around 100,000 particles on the wafer to be a good starting point, as the proportion of incorrectly identified particles increases significantly with higher particle numbers. In the case of lower particle numbers, the number of INPs on the wafer may be too low depending on the INP fraction, in the case of higher loadings, the fraction of ambiguous positions increases.
To obtain the best results, we recommend to determine the particle concentration in the atmosphere in parallel to the collection and then perform a quick analysis of the wafers in FRIDGE to calculate the proportion of INPs in the total aerosol. Based on this proportion the sampling parameter can be adjusted to balance a sufficient number of INPs on the wafer but prevent an overload."

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

We added a definition of the identification rate.

"The identification rate is defined as the proportion of ice crystal coordinates examined by SEM to which an unambiguous INP could be assigned."

-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.

Yes, this value is based on the 14 analyzed samples from the JFJ campaign. We added a few words to clarify this.

"The average INP identification rate for the JFJ samples (based on 14 analyzed samples) was calculated to be 30%.). The large variation from 13 % to 50 % for the individual samples is an expected consequence of the different wafer loadings of the individual samples, as well as of the partly low counting statistics."

-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 discussed these values.

Generally, it can be expected that the number of positions with multiple particles depends on the total wafer loading, as previously mentioned in the text and seen from the model: If there are few particles on the wafer, there will be few multiple particle positions. A greater number of particles on the wafer will increase the number of multiple particle positions.
However, a correlation with sampling time/volume, or particle concentration / number of particles on the wafer is not a straight forward task. Since we have a loss of volatile components, the airborne particle concentration may not be directly relevant for the number of particles deposited on our wafer. The spatial distribution of the particles on the wafer also plays a role. Normally, the particles should be very uniformly distributed over the entire surface of the substrate. However, some areas of the wafer surface may have a heterogeneous distribution of particles. In such areas, the chance of getting a multiple particle position is higher than on the rest of the substrate.
We have checked possible correlations for the samples from the JFJ campaign and did not find a good correlation for the total particle concentration for particles > 0.1 µm and > 0.5 µm.

"In addition, the study identified the presence of multiple particles at 45 % of the locations, ranging from 7 % to 81 % for the individual samples. Although a correlation of multiple particle positions with the total number of particles on the substrate surface is suspected, such a correlation is not obtainable directly, as the particle distribution on the wafer and the possible loss of volatile components (see Sect. 2.6) have an impact."

-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?

The presence of a blank position can be attributed to several factors. Firstly, particle shift, defined as the displacement of particles during the process of ice growth, can result in a blank position if the particle ceases to be at the center of the ice crystal. Second, erroneous identified coordinates can also lead to a blank position if the crystals did not grow symmetrically and the origin was not identified correctly. Third, the limitation of the search radius to 50 µm can also be a contributing factor. In general, the optimal search radius in SEM increases with increasing uncertainties. In contrast, this radius decreases with increasing wafer load. As previously mentioned, the value of 50 µm represents a compromise between the consideration of the uncertainties and the uniqueness of the position. Therefore, it is possible that a position is classified as a blank, but would have a clearly assignable INP at, say, 80 µm. In such cases, extending the radius to 100 µm might prove beneficial, if the sample loading is sufficiently low.
We have added the missing third point to the description of a blank position.

"(3) In the absence of a particle within the 50 µm radius, these blank positions are disregarded. A blank position may be the consequence of possible particle shift during the processing in FRIDGE (discussed in Sect. 2.3), or the result of an erroneous calculation of the ice crystal origin (discussed in Sect. 2.4). As it can be seen from the model simulations, an incorrectly selected search radius can also lead to a higher number of blank positions. In cases where the substrate loading is low, it may be beneficial to increase the search radius in the case of a blank position in order to increase the number of identified INPs."

The influence of the different factors for a blank position on the results is discussed in the section on identification rates.

"The remaining 25 % (ranging in the extremes from 2 % to 66 %) were found to be blank positions, for the reasons discussed above. In case of a blank position due to an uncertain ice crystal position in connection with the search radius restriction, the overall result remains representative. No bias is expected in the relative contribution of individual particle classes. In the case of particle shift being the reason for a blank position, there is a potential for a bias in particle frequencies or size distribution, if the potential for a particle to shift would correlate with particle class or size. Similarly, as volatile components and thin films cannot be detected in the electron microscope reliably (Sect. 2.6), a blank position might be detected here, although they could have triggered ice formation in FRIDGE."

-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?

The uncertainty of our classification can be derived from the counting statistics in the supplement (Tab. S1 – number referring to the new manuscript). Judging from the statistics with 95% confidence interval, quantitative statements about the relative frequency of a particle class are reliably possible starting from approx. 10 particles per group. For groups with less particles, the statistical uncertainties become larger.
In our specific case, the counting statistics with 95% confidence interval (Table S1) show that for the SDE with only 70 analyzed particles in total, a quantitative statement is only possible for the main component (Al-rich / aluminosilicates), whereas for the non-SDE period with 129 analyzed particles, a statement about 4 groups (aluminosilicates / Al-rich, carbonates, C-rich and mixtures) is possible due to the higher number of identified particles.

"Based on the uncertainties and assumptions discussed, many positions and potential INPs are excluded from further analysis. As a result, the number of identified INPs per sample is limited and typically low in contrast to the grown ice crystals in FRIDGE. These INPs, however, are accurately identified as we know that ice formation has taken place on the substrate at their position. This allows for conclusions on the analyzed INPs within the limits of their counting statistics and depending on the significance level, even for a small number of identified INPs. From the statistical calculation in Tab. S1 with a 95% confidence level, a limit of approximately 10 particles per group can be derived to make a reliable quantitative statement, for groups with less particles the uncertainties

become large. Further considerations of the limits of certain statements must depend on the particular statement and should employ common statistical approaches for counting statistics and compositional data."

-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 µm was the size of the grid on the FRIDGE image, please add some scale to avoid confusion.

The figure has been improved as follows:
A caption has been added to the FRIDGE image stating that it is a FRIDGE image with ice crystals. In addition, the ice crystals have been numbered so that they can be directly linked to the corresponding identified INPs. A legend for the electron microscopy result has also been added for better understanding, as well as a scale.
The axis labeling of the spectra has been made consistent, a y-axis has been added. The figure is intended to give the reader an overview of the method, so the images and spectra have a more symbolic character.
Our samples are collected on Si wafers, so the Si peak is ubiquitous in our spectra, and it is extremely high. Therefore, the peak is cut off, which is common practice to show relevant data.

-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?

In the case of an INP consisting of a main particle showing small inclusions or accretions (Fig. 7 – number referring to the new manuscript), the classification is determined by the main particle. However, if the INP obviously consists of several components lying next to each other (Fig. 6 INP4 – number referring to the new manuscript), the particle is considered as a mixture.
We added a sentence to clarify, how we proceed with mixed particles.

"In case of a main particle with small inclusions (as shown in Fig. 7 and Fig. 10d, for example) the composition of the main particle determines the classification. If a particle clearly consists of several individual components (see Fig. 6 INP4, for example), it will be classified as a mixture."

-L.312: "surface properties" what kind of properties?

In general, we can identify surface properties such as cracks, smooth or rough structures and edges in SEM. Even if we cannot say exactly at which point the ice nucleation starts. We have decided to mention them here anyway as a general possibility of SEM, even if they are not the focus of this study.

-L327: "chemical characterization" why chemical or not elemental composition?

In general: When referring to the results of the EDX analysis, we have chosen to use the term "elemental composition" instead of "chemical composition", as this is the result obtained from the analysis for each individual INP. This is done to illustrate that, from this point on, we assume that the detected elements form a chemical compound (and not for example a random mixture). Therefore, when we refer to the different classes of INPs, we prefer stick to 'chemical composition'.

-L345: "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 (CO_3)?
-L378:" 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?

In our spectra, the peak area, even after background subtraction, is not directly proportional to the element concentration, as the peaks are – besides the element contents -impacted by different excitation rates, X-ray absorption and secondary fluorescence in the particle matrix. This is particularly the case for elements with low X-ray energies < 1 keV. The problem eases at higher energies. For example, quantifying the element ratios for the Ca-sulfate from Fig.10 (new manuscript) leads to a 1:1 ratio (atom%) of Ca and S.

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

Changed as requested, added the zero.

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

The images are now labeled and referenced in the figure caption. We have also added a reference to distinct pictures in the text in certain places.

"Figure 10: Overview of representative EDX spectra and corresponding SEM images for the defined INP classes grouped as mineral components (aluminosilicate / Al-rich (a), Ca-carbonate (b), silicon dioxide (c)), carbonaceous particles (biological particles (d), soot (e), C-rich (f)) and other particle classes (Ca-sulfate (g), metal oxide (h), mixtures (i))."

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

The relative humidity is RH = 99 % for each of the three activation temperatures shown. It is mentioned in the figure caption (Fig. S3 - number referring to the new manuscript).

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

With this figure, we would like to give the reader a brief overview of the FRIDGE data from the campaign. As the graph already contains a lot of data points and information, we believe it would be too confusing if error bars were also shown for each individual data point. We have therefore decided to only mention the error as a value in the text. For clarification, we added the error also to the figure caption. The figure has been moved to the supplement (see following comment on Fig. 8).

"Figure S3: INP concentrations (deposition nucleation / condensation mode freezing) calculated from the FRIDGE measurements at RH = 99 %. The concentration for each sample is calculated on the basis of one measurement. For days with more than one sample, an average value was calculated. Days with analyzed samples are indicated by triangles and the corresponding sample numbers. The 5-day running average concentration is shown by the dotted lines (the figure is adapted from Weber (2019)). The error of the concentrations given here is 20% approx., based on the relative error of the counting uncertainty for individual measurements from Schrod et al. (2016)."

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

For the campaign, the collection volume of the individual samples ranged from 115 to 306 liters.
Since particularly low INP concentrations were expected in the free troposphere at JFJ in advance, special attention was paid to the cleanliness of the sample substrates. The blank value for the campaign was therefore particularly low (less than 3 ice crystals at -30°C).
When calculating the INP concentration, the blank values are directly subtracted from the number of grown ice crystals. The concentration shown in Fig. 10 is already blank-corrected.

-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.

We changed the colors a bit and increased the size of the datapoints. However, in order to focus even more on the coupling, we have decided to move the figure to the supplement.

-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?

The identification rate indicates that a unique INP could be assigned to 30% of the analyzed positions. When analyzing the JFJ samples, only a fraction of the area, a defined section around the center of the wafer (square of 2x2 cm) was inspected.

"Overall, based on the parameters described in Sect. 2, we were able to clearly identify and characterize the associated INPs for 200 ice crystals, which corresponds to 30 % of the ice crystals positions analyzed (Fig. 12b). For the remaining 70 % we were unable to make a statement. While the multiple particle positions have no effect on the proportion of particle classes (Fig. 11) or size (Fig. 12a), the blank positions can cause a bias (discussed in Sect. 2.5.3). In this campaign, only a square with a side length of 2 cm in the center of the substrate was analyzed

by electron microscopy. Since the area analyzed in SEM corresponds to roughly half of the area considered in FRIDGE, the 200 INPs identified represent about 15% of the total sites activated in FRIDGE. This limitation has no influence on the individual chemical fractions or on the size distribution of the INPs, as the area was chosen arbitrarily and a homogeneous distribution of the particle groups on the wafer can be assumed."

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

Changed to "chemical composition of the INPs".

The same applies here as described above: When referring to the results of the EDX analysis, we have chosen to use the term "elemental composition" instead of "chemical composition", as this is the result obtained from the analysis for each individual INP. This is done to illustrate that, from this point on, we assume that the detected elements form a chemical compound (and not for example a random mixture). Therefore, when we refer to the different classes of INPs, we prefer stick to 'chemical composition'.

-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 proving all spectra in the SI?

All spectra can only be read and edited with the internal Oxford Aztec EDX software. To publish a spectrum, it must be exported, converted and edited individually. In our opinion, the effort involved is too great to publish a complete data set in the supplement.
If the data is required for a specific purpose, it can be requested. This allows us to customize the data to specific interests.

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

Changed as requested.

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

We added the explanation how the projected area diameter is calculated.

"Therefore, the particle is regarded as an ellipse and the dimensions of its major and minor axis are determined in order to subsequently calculate the diameter of an equivalent circle, which is referred to as d_{pa}."

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

We included the uncertainties in the supplement (Tab. S2 – number referring to the new manuscript). It is now stated at the beginning of the case study, how much INPs were analyzed and they are related to the total number of INPs grown in FRIDGE (Sect. 3.2). For a better understanding, we have added a second graph (Fig. 12 b – number referring to the new manuscript) which shows the proportion of the ice crystals examined that correspond to the graph in Fig. 12a.

„Overall, based on the parameters described in Sect. 2, we were able to clearly identify and characterize the associated INPs for 200 ice crystals, which corresponds to 30 % of the ice crystals positions analyzed (Fig. 12b). For the remaining 70 % we were unable to make a statement. While the multiple particle positions have no effect on the proportion of particle classes (Fig. 11) or size (Fig. 12a), the blank positions can cause a bias (discussed in Sect. 2.5.3). In this campaign, only a square with a side length of 2 cm in the center of the substrate was analyzed by electron microscopy. Since the area analyzed in SEM corresponds to roughly half of the area considered in FRIDGE, the 200 INPs identified represent about 15% of the total sites activated in FRIDGE. This limitation has no influence on the individual chemical fractions or on the size distribution of the INPs, as the area was chosen arbitrarily and a homogeneous distribution of the particle groups on the wafer can be assumed."

Technical corrections

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

Changed as requested.

"As a routine, the wafers are measured in 12 cycles combining three temperatures (T = -20 °C, -25 °C, -30 °C) and four relative humidities (RH) (RH = 95 %, 97 %, 99 %, 101 %)."

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

Changed as requested.

"It can be assumed that this coordinate represents the position of the corresponding INP, since an approximately radially symmetric ice crystal growth can be observed in the range of the selected activation conditions in FRIDGE."

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

Changed as requested.

These positions are excluded from further analysis.

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

Changed as requested.

"A Quanta 200 FEG environmental scanning electron microscope (ESEM) by FEI (Field Electron and Ion Company; Eindhoven, Netherlands) coupled to an energy dispersive X-ray detector (EDX) (EDAX, AMETEK, Tilburg, Netherlands) was used for analysis."

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

Changed as requested.

"The metal oxides are characterized by the presence of oxygen and a corresponding metal (except Al, which is assigned to the previous aluminosilicate / Al-rich particles group)."

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

GAW stands for "global atmosphere watch" which is a program of WMO which focusses on the monitoring and understanding of atmospheric composition. As the information is not necessary for the reader to understand the results, we removed the GAW in this case.

"Aerosol sampling for the FRIDGE experiment was conducted downstream of the total inlet (Lacher et al., 2021)."

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

This statement refers to Fig. 2, so yes, we have shown that. However, we adapted the wording.

"It has been shown, that our position identification algorithm works reasonably well, with a negligible number of incorrect positions due to condensation or coalesced ice crystals."

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

Changed as requested.

"Although the method has some drawbacks and uncertainties, it enables high accuracy in the identification and in this way physico-chemical characterization of individual INPs."