Author response to Reviewer 1

First of all, we would like to thank the reviewer for reading and commenting on our manuscript. The comments and remarks helped us a lot to focus more on the methodological aspects and to improve the structure of the manuscript.

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.

General comments

In this study, the authors present an offline method for characterizing ice nucleating particles (INPs) by coupling an offline diffusion chamber, the FRankfurt Ice nucleation Deposition freezinG Experiment (FRIDGE), with a scanning electron microscope (SEM). The approach is potentially useful to provide comprehensive information of INPs, including their chemical composition, morphology, and size, and to improve the parameterization in the corresponding models. However, the advantages of combining the two instruments are not well demonstrated from the case study at JFJ. The FRIDGE-SEM coupling technique is not new and no convincingly new findings are shown in this study compared to the former studies at the JFJ. The methodology needs more details as intended to, in particular the coordinate system that allows for recovery of the particles, which is critical to determine whether particles are original or processed INPs. Statistical significance is also a big concern. It is unlikely to use the data present in this study to evaluate the INP-type-specific parameterizations in the model. In addition, the manuscript is not very well structured. The potential contribution of this study is within the scope of AMT. However, the current manuscript is behind publication quality due to the reasons as mentioned above. Therefore, I recommend that substantial revision needs to be done before considering publication.

We have revised the entire manuscript (see our responses to the major and minor comments below as well as our responses to the other two reviewers for details). Structural changes have been made, to focus more on the method. The entire method section has been revised to make it less manual-like, as it was suggested by the reviewers. The case study discussion has been shortened, in favor of a more concise and focused methodological manuscript better fitting the scope of AMT.

The coupling of FRIDGE and the SEM is now described in more detail and in a more structured way in the methods section, where advantages, drawbacks and uncertainties are discussed and a general overview of the potential of the method is provided.

The case study in this manuscript is intended to illustrate that the method presented provides reliable results for the main INP groups, despite relatively low identification rates, and not primarily to present new atmospherically relevant results. The discussion of the results has been adapted to emphasize comparability with other studies. Despite the small number of analyzed INPs, we have decided to show a comparison of air masses before and during a Saharan dust event, as the potential of the unambiguous particle assignment to the corresponding ice crystals becomes obvious here.

We do not claim that the presented results of our case study are sufficient to be used for model evaluation, we just want to express that this type of results could be used for a closure study as suggested to Burrows et al. (2022).

Major Comments

The novelty of this study is supposed to be the technical details and direct measurement of INPs. However, it needs more details to convince the readers that the individual particles can be fully recovered after measurement cycle of the FRIDGE. Additional controlled experiments may need to be done and shown to prove that hypothesis. Otherwise, the SEM measurement would be on the IRs rather than original INPs.

Based on the definition by Cziczo et al. (2017) the particles activated in FRIDGE are INPs because they were activated under defined conditions after the collection of the total aerosol. In the electron microscope, we are able to locate and analyze these INPs at the positions of crystal origin.
Of course, in this context one could also speak of ice residuals (IR), since the particles have already been processed in FRIDGE. IRs are defined as particles remaining after the collection of atmospheric ice crystals and subsequent evaporation of the ice phase. In this case, there is a risk that additional particles deposited on the surface of the original ice crystal are subsequently identified as ice forming particles. This problem does not exist in our method. This is why we refer to the analyzed particles as INPs.

"*Based on the definition by Cziczo et al. (2017) we refer to the identified particles as INP, as they were activated under defined conditions after the collection of the total aerosol and not sampled as ice crystals. Therefore, we are able to investigate truly activated particles in contrast to methods analyzing IRs, which face challenges in order to distinguish between IRs and scavenged particles. However, some of the INPs analyzed with SEM may have undergone changes (see Sect. 2.6) due to the measurement procedure in FRIDGE, but we assume that these changes are of minor importance for the main INP classes that we can analyze with this method.*"

It is good that comparisons with previous studies at JFJ are discussed. However, the authors may want to add more discussion on the differences and emphasize the new findings.

In our opinion, a case study in a methodological paper should exemplarily showcase the capabilities of a measurement system by presenting plausible results that, in the best case, agree to previously published literature. As shown in the revised manuscript the results indeed are in good agreement to those from Eriksen Hammer et al. (2018) and Lacher et al. (2021) that were obtained from the same campaign, as well as to those of Worringen et al. (2015), Kamphus et al. (2010) and Ebert et al. (2011) measured at JFJ but a few years prior. For a paper that focuses on atmospheric measurements and their implications, we agree with the reviewer that it is important to discuss differences and, above all, to focus on new findings, however, we think that such an in-depth discussion would be beyond the scope of an AMT paper. In fact, to strengthen the methodological focus of the paper, we have streamlined the case study section.

The results were based on 200 individual particles from 5-week measurement, and only half of the total analyzable area of each wafer was analyzed by SEM. The author should evaluate the statistical significance, discuss more about the representativity, uncertainty and limitations.

Of course, 200 INPs for a period of more than 5 weeks is comparatively low. Therefore, we do not claim that our results are representative of an average composition of INPs over the entire campaign period. This is now also noted in the manuscript.

"Although the number of identified INPs appears comparatively low for a campaign period of five weeks, these INPs were identified with a high degree of reliability (Sect. 2.5.2). The small number of particles identified bears the risk that individual, time-limited variations occurring randomly during the sampling periods may influence the resulting total composition to a certain degree. It should

therefore be noted that the results presented below may not comprehensively reflect the main composition of the INPs over the entire campaign period. Nevertheless, it can be shown that the method provides valid results for the main groups of INPs (see confidence intervals for Fig. 9 in the supplement (Tab. S2)).”

However, by comparing air masses before and during a Sahara dust event (see Fig. 9 in the revised manuscript), we were able to show that it is possible to identify important trends in INP-relevant groups. This illustrates the great strength of our method, which has low identification rates, but identifies the INPs with high accuracy and thus still delivers credible results. Confidence intervals for the chemical distribution are given in the revised supplement (Tab. S2).

The structure of this manuscript needs revision. The introduction, method and results sections have several overlaps, which need to be improved. E.g., in the introduction section, L 126 to 130 belongs to method part; Section 2.6 Chemical classification should be merged into Section 3.4 INP chemistry or go to supporting information; Section 3.1 Sampling site needs to go to method and/or introduction; Section 3.3 Method evaluation needs to go to method section.

We have revised the structure of the paper and created a distinct separation between the general methods section and the results of the case study. With regard to the points mentioned, we have proceeded as follows:

L126 to L130 from the introduction section was moved to the method part as an introduction.

Section 2.6: We have decided to leave this section (renamed as "individual particle analysis") in the methods section and have adapted it accordingly. It no longer describes the specific particle classes found at the JFJ but defines a general classification scheme and discusses strengths and limitations of the individual particle analysis.

Section 3.1: For a paper focusing on the results, we would agree with the reviewer. But in our opinion, in this case, the sampling site description belongs to the case study, because the method part describes and discusses the coupling procedure and the sampling site is part of the case study.

Section 3.3: As the identification rates are highly dependent on the total wafer loading, which is influenced by the aerosol concentration and activated fraction during the sampling time, the values for the JFJ campaign are not universally valid. We have therefore not included these values in the methods section. However, we understand the reviewer's point, which is why we have decided to include the identification rates with the corresponding restriction in the method section (Section 2.5.2 "INP identification").

“The number of INPs that can be unambiguously attributed to an ice crystal origin is significantly influenced by the total wafer loading, which is determined by the sampling parameters (e.g., flow rate, sampling time, deposition efficiency) in combination with the aerosol concentration present. However, 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. This ratio is variable and usually unknown prior to measurement. As a result, the amount of atmospheric aerosol and the proportion of INPs deposited on a wafer are highly variable. This variability is also seen in the identification rates, which is why it would be misleading to give an average identification rate for the method presented. However, a specific identification rate for the case study conducted at the high-altitude research station Jungfraujoch (Sect. 3) can be given here as a guideline. The average INP identification rate was calculated to be 30% (ranging from 13% to 50%). Furthermore, the study identified the presence of multiple particles at 45% of the locations (ranging from 7% to 81%), while the remaining 25% (ranging from 2% to 66%) were found to be blank positions.”

The comparison between the different ice crystal counting methods (formerly Fig. 5) was also moved to the method section (Section 2.4 "identification of ice crystal positions") and replaces the graphical representation of coordinate determination (formerly Fig. 2), which has been moved to the supplement.

Minor Comments:

L33: Please remove "e.g." from the citation and do it through the entire manuscript.

We have removed the "e.g.," in many cases. In some cases, however, we have left it to indicate that the reference cited is just an example.

L41-42: What is the temperature range you mentioned? Please add the values and explain why the main focus has been on such range.

The temperature range for heterogeneous nucleation in mixed phase clouds is 0°C > T > -38°C. At temperatures colder than -38°C spontaneously homogeneous freezing might dominate the formation of ice. In the case of homogeneous freezing no INPs are needed to exceed the energy barrier. That is why the main research focus for INPs has been on the temperature range between 0°C and -38°C.

However, these sentences have been deleted in favor of a more focused introduction with less of a review character.

L44: Quantify the "small fraction". How much%?

The sentence has been adapted and now reads:

"Only a small fraction of the total aerosol can act as INPs and their concentrations can show variations of several orders of magnitude in space and time (DeMott et al., 2010; Kanji et al., 2017)." Values for the proportion of INPs in the total aerosol are given in the two references.

However, later in the Introduction (L96-97) we give values:

"… 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),…"

L46 & 51: Change "history within the atmosphere" to "atmospheric processing"

Changed as requested. The entire section has also been shortened. The new sentence now reads as follows:

"In addition to the prevailing environmental conditions i.e., temperature and humidity, the potential for an INP to become activated is dependent upon individual particle properties (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)."

L60: Mineral dust is a good INP, rather than an important factor in ice nucleation.

Changed as requested.

"Mineral dust, which is emitted from arid and semi-arid regions and is globally distributed in the atmosphere (Perry et al., 1997; Ansmann et al., 2003; Schepanski et al., 2018), is a good INP at temperatures below -15°C (Hoose & Möhler, 2012)."

L76-93: Please consider remove or shorten this part. Lab, field and model studies are the main approaches for atmospheric science. This is very basic and general, not specific for ice nucleation studies.

We have removed the entire section. The references for laboratory and field experiments have been moved to the following section.

"Although there is a variety of methods to determine the INP concentration in the laboratory (Hoose & Möhler, 2012; Hiranuma et al., 2015; DeMott et al., 2018; Hiranuma et al., 2019) and in the field (Wex et al., 2019; Schrod et al., 2020b; Brasseur et al., 2022; Lacher et al., 2024), only a few of them are simultaneously able to report on the chemical characteristics of individual ice-nucleating particles."

L103-104: The sentence is unclear. Please revise.

Revised sentence: "In another approach, the particles are activated under defined conditions in an online reaction chamber (e.g., Rogers, 1988) after the collection of the total aerosol."

L105-106: It is unclear why the IR and INP are in the brackets. Please add more descriptions.

The entire section on the collection of INPs/IRs has been rewritten and shortened. As a result, the entire definition of INPs and IRs (including this sentence) has been removed.

L111: What are the problems? Please elaborate.

Online systems may have problems to resolve low concentrations of INPs as they have typically a lower sampling flow, resulting in low INP counts in the range of background counts. Offline methods, on the other hand, can enrich the number of INPs on the sample substrates due to higher sampling flows and longer collection times.

L116-122: You might want to emphasize more on the advantages of EM over online measurement technique. For example, the morphology and quantitative results that SPMS generally does not provide without reference instrument. Note that SPMS can provide information of mixing state, which is not only obtained by the EM.

We removed the mention of the mixing state in the section about EM.

L131: I am not convinced that the particles characterized by SEM are INPs rather than IRs.

The particles originally collected and activated in FRIDGE are INPs, since they were collected as total aerosol and not as ice crystals. The processing in FRDIGE (repeated activation/evaporation) can undoubtedly lead to changes in particle properties in some cases, which is why one could also speak of IRs in EM. However, according to our understanding, these changes are more likely to affect the soluble/volatile components, which are not known to contribute significantly to ice nucleation in the considered temperature range. Since our method does not allow us to detect the small volatile components well, we assume that the majority of particles which can be analyzed with our method do not undergo serious changes during the activation/evaporation processes. Therefore, we stick to the definition of Cziczo et al. 2017, according to which we collected and activated INPs. In addition, the coupling allows for investigating truly activated particles in contrast to methods analyzing IRs, which face challenges in order to distinguish between IRs and scavenged particles.

L133-148 Figure 1: It is unclear regarding the second and the third pictures, please consider adding B1, B2 and B3 for second and third pictures regarding the FRIDGE, C1, C2 and C3 for SEM. Please add the missing legends for the bars and pie chat. Please add missing units for the x-axes on the

EDX spectra and the time series of INP conc., respectively. Please increase the resolution of the pictures, especially the scales and labels which are too vague for current version.

The desired labels (B1, B2 and B3 for the FRIDGE as well as C1, C2 and C3 for SEM) and the missing labels for the pie chart and the bars have been inserted. The labels for the pie chart and the bars have been kept general (TA, TB, TC and Class A, Class B, …) to make it clear that this is only a sketch of the method and that no real results are shown. The missing units have also been added and the scales and the font size of the labels have been increased.

L174: Change “at” to “in”.

Changed as requested.

L205: Will measurement cycle change e.g., morphology or properties of particles? If no, you need to give the proof.

Please, see comment on INP discussion above. Of course, the morphology, especially for soluble particles can change during the activation/evaporation process. Since most of the particles we identified as INPs consist of non-soluble components, we believe that this does not significantly affect the results.

L204: Change “electron microscopy” to “EM”. The full name only needs to be mentioned when shown for the first time in the manuscript (from introduction section).

Changed as requested.

L223: What is ImageJ? Please explain.

ImageJ is a free image processing software. We added this information to the text.

“The ice crystal positions are identified by image analysis using the internal particle analyzer of the free image processing software ImageJ (Schneider et al., 2012), with a minimum size of 30 pixels proven to be useful.”

L231: Consider changing “time-consuming” to “labour-intensive” or other more appropriate word. Please revise and give the approximation of the duration of such analysis.

The corresponding section has been rewritten for the purpose of a less manual-like writing style, as it was requested by the reviewers. The wording is no longer used in the new section.

L247: Give full name for “FEI”

FEI is the official company name. It stands for “Field Electron and Ion Company”.

“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.”

L254: Remove the full name of SEM.

Changed as requested.

L267: How much is the uncertainty? Please give the value.

This uncertainty is based on the calculation of the ice crystal origin and the coordinate calibration. We added a comment on the uncertainty for the ice crystal calculation in Section 2.4.

“Nevertheless, a potentially imperfect radial symmetry of the ice crystal growth, coupled with the restricted resolution of the FRIDGE images (20 x 20 µm), may result in an uncertainty in the calculation of the ice crystal origin. As the size of an ice crystal increases, the probability and extent of such a non-symmetrical growth also increases. The quantification of this uncertainty proved to be difficult, as it depends on the symmetry deviation present. 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.”

The coordinate calibration uncertainty is mentioned in Section 2.5.1.

“Due to the limited resolution of the FRIDGE images of about 20 x 20 µm, the calibration has of course an uncertainty in the same order of magnitude.”

Regarding the accuracy of the coordinate, the following is stated in the INP identification section (Section 2.5.2):

“Each ice crystal position, *based on a real grown ice crystal, is inspected by SEM to identify the presence of particles. Given the uncertainties associated with the ice crystal identification process (Sect. 2.4) and the coordinate calibration (Sect. 2.5.1), it is crucial to consider not only the exact calculated coordinate but also the surrounding area. This area must take into account the aforementioned uncertainties and, at the same time, limit the probability that several particles will be observed in the scanned area. In this context, a radius of 50 µm has proven to be useful. While the previously discussed uncertainties may suggest a lager radius to be beneficial, in fact, the high substrate loading often proves to be the limiting factor.”*

L294: How much smaller? Please give the value.

The section has been rewritten:

“The number of INPs that can be unambiguously attributed to an ice crystal origin is significantly influenced by the total wafer loading, which is determined by the sampling parameters (e.g., flow rate, sampling time, deposition efficiency) in combination with the aerosol concentration present. However, 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. This ratio is variable and usually unknown prior to measurement. As a result, the amount of atmospheric aerosol and the proportion of INPs deposited on a wafer are highly variable. This variability is also seen in the identification rates, which is why it would be misleading to give an average identification rate for the method presented. However, a specific identification rate for the case study conducted at the high-altitude research station Jungfraujoch (Sect. 3) can be given here as a guideline. The average INP identification rate was calculated to be 30% (ranging from 13% to 50%). Furthermore, the study identified the presence of multiple particles at 45% of the locations (ranging from 7% to 81%), while the remaining 25% (ranging from 2% to 66%) were found to be blank positions.”

L302: “appearance”, do you mean morphology?

Yes, we mean morphology. We changed the wording.

“Therefore, the morphology of the particles may have undergone alterations.”

L305: Consider changing “excellent” to “efficient”

Changed as requested.

L374 Figure3: The scales and labels are too vague. Please increase the resolutions.

Changed as requested. The y-axis of the EDX-spectra are not important, as they show only the counts, which are dependent on the EDX sampling time and the current of the electron beam.

L437-438: Please separate the samples into cloudy and clear sky cases and discuss accordingly.

As requested by the reviewers, the case study section was shortened to emphasize the focus on the methods section. To emphasize the potential of the method, we decided to show a separation for airmasses influenced by an SDE and prior to that event. Although the suggestion to separate the samples into cloudy and clear sky cases is certainly interesting, we fell like that this would go beyond the scope of an AMT paper.

L440-444 Figure 6: Cloudy and clear sky cases should be separated.

As previously mentioned, we have decided to distinguish the SDE air masses from the air masses prior to that event, as this illustrates the potential of the method. Despite low particle numbers, the high accuracy in identifying the INPs allows significant differences between different air masses to be recognized.

L634-636: Consider archiving data in a reliable public data repository.

The complete data set is available for the community and can be accessed by request to the corresponding authors.

Author response to Reviewer 2

Firstly, we would like to thank the reviewer for reading our manuscript and the comprehensive review, which helped us to describe the method in a more focused way.

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.

The manuscript reviews how ice nucleating particles can be located on a wafer and their composition and size analysed using an electron microscope. The use of the method is exemplified on a set of wafers collected at Jungfraujoch in 2017. The coupling of the ice nucleation chamber FRIDGE with EM analysis is very suitable for the task of gaining information on the abundance of a specific category of particles active as INP at a certain temperature. However, the methodology and detail of coupling FRIDGE with EM has been discussed in previous papers and it is not made clear what novel information is provided in the current manuscript. Concerning the methodology, it seems not to go beyond what is already published in Schrod et al., 2016 and He et al., 2023.

Schrod et al. (2016) essentially evaluates FRIDGE as a method for determining INP concentrations based on the deposition nucleation/condensation freezing modes, without consideration of the coupling process. He et al. (2023) provides a brief overview of the coupling, with the primary focus being on the presentation of results.

This technical paper presents an overview of the FRIDGE measurement, outlining the critical aspects that must be taken into account to ensure the reliability of the results. It also provides a comprehensive description of the techniques employed for ice crystal detection and electron microscopic analysis. By providing a detailed description of the method, it is possible to demonstrate both the strengths and limitations of the method and to establish a guideline for the interpretation of the corresponding results. The method has been in development for several years in our working groups, and the paper by He et al., (2023) is based on our method, although the coupling of the devices (crystal detection, etc.) does not correspond exactly to our method. From the paper it is not clear, which exact method is used to identify the ice crystals and to find the corresponding positions of the ice crystal origin in the SEM.

The JFJ case study is a valuable dataset by itself, but the attempted validation of the FRIDGE-EM coupling by comparing to different techniques that investigated the INP composition at different activation temperatures on JFJ is not convincing. As the authors note themselves at best only a rough comparison can be made.

We agree that it is often challenging to make direct comparisons between INP measurements obtained from different devices, as the different collection processes, specific activation conditions and different classification schemes can have a significant impact on the resulting data. Nevertheless, we firmly believe that the comparison for the main INP classes, which is made in the revised manuscript, is meaningful enough to demonstrate that our method yields reliable results.

Listed in the comments below are several inaccuracies and inappropriate references.

The line of explanations should also be structured clearer. On several occasions, statements are made that are not comprehensible and are explained only by information given later in the text. Leading with the necessary information and explanations before a conclusion or result, would make it easier to follow.

We understand that in some places it may have been a little difficult to follow our line of thought. The whole manuscript has been restructured. All methodological elements are now part of the method description and discussion. Only the INP-specific results (concentration, chemistry, size) for the campaign are shown in the case study section.

It is irritating that the author state on couple occasions that a detailed analysis is not possible or feasible, but the analysis is then done anyway in the following.

Maybe at some points, reading the comparison of the case study results to other studies was a bit imprecise and thus lead the reviewer to believe that a deeper analysis is generally not possible. The idea behind our structuring of the results section (chemistry and size of the INPs at the JFJ) was to first present the results, followed by a *comparison with other data from the same campaign and then a comparison with previous studies at the same location. Since the comparisons were carried out study by study, we also saw the need to point out differences / non-comparable points for the purpose of providing a complete picture.*

The structure of the result discussion (where most of these statements were made) was changed. The results are now always compared directly after the individual particle class description. This allows us to highlight comparable results from different studies. The statements are now more general, and therefore a comparison is feasible.

Nevertheless, the above-mentioned limitation of comparability is mentioned in a general statement.

Section 3.3 (INP chemistry):" It is generally difficult to make direct comparisons between the results of different INP/IR measurement techniques, as the results can vary significantly depending on the sampling configuration, ice nucleation activation conditions, and the classification schemes used for each instrumentation."

Section 3.4 (chemically-resolved INP size distribution): "The comparison of such INP size distributions with chemical information from different methods is difficult, since in addition to the influencing factors discussed in Sect. 3.3, a possible size selection or limitation of the sampling process, and different techniques of particle sizing may also play a role."

Because the manuscript lacks novelty, rigor and structure, I recommend major revisions before consideration for publication.

We disagree on the point, that our manuscript lacks novelty, as this paper describes the coupling between FRIDGE and SEM for the first time in detail (see first comment above).

A lot of structural changes have been made as recommended by all reviewers (e.g., method evaluation from the case study moved to the method section).

Specific comments:

Title: The analysis of particle morphology is not discussed in the manuscript. Consider adjusting the title accordingly.

We decided to stick to morphology in our title, because the morphology of particles was used to classify them. (revised manuscript Sect. 2.6). In addition to that, the focus of this paper should be the method description. And therefore, it is possible to analyze the morphology of individual INP with this method, even if we didn't focus on this aspect in our case study.

"The analysis by SEM and EDX is an efficient method for characterizing INPs in detail, as it provides information on elemental composition and distribution as well as on morphology and surface properties. The morphological information can be used for source apportionment (e.g., biological particles, soot, spherical particles from high temperature processes). With this detailed information, it is possible, for example, to determine the mixing state of a particle (see Fig. 3 and Fig. 4)."

Abstract

Line 29: Specify how the results can be used to evaluate parametrizations.

Such results may be useful as part of a closure study, which combines size-resolved aerosol composition measurements, particle class dependent INP parametrizations and INP measurements, including size and chemistry information in addition to their concentration (see Burrows et al. 2022). For more details see also comment on L85-89.

Introduction

The introduction should be shortened and focussed on motivating the presented methodology and be less of a review of the subject area in general.

We now focused more on the method motivation. Therefore, we shortened / re-arranged some sections in the introduction (e.g., removed the part on laboratory/field experiments and modelling (L76-93 in the original manuscript)). The motivation of our method by analyzing particles at the position where ice growth has occurred compared to the collection of ice crystals and the associated risk of artifacts, e.g., due to previous riming processes, was clarified. However, we believe it is essential to provide a comprehensive introduction and overview of INPs, as it is the basis for our method.

"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. Even with an accuracy of 99.9% for INP separation, this would mean that for every correctly separated INP 1000 non-INP particles would be separated incorrectly, when an INP to total aerosol ratio of 1/10^6 is assumed. In this way, no conclusions about the chemistry of the INP would be possible at all, since there is no way to distinguish particles that have been falsely separated as INP from real INP 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 INP/IR fraction by three different methods in Worringen et al. (2015)."

Instead of repeatedly citing chapter 1 and 8 from the 2017 AMS reviews of Kanji et al. and Cziczo et al., it would be more helpful to cite specific references to the individual topics.

We think that it makes sense for general statements to cite the reviews. When there were more specific references that better reflect the statements made, we now cite those studies instead. For example, in line 59-60 we now cite Archuleta et al. (2005) and Yakobi-Hancock et al. (2013) when discussing metallic particles.

"Their efficiency to activate as INPs depends on the type of metallic cation as well as on the oxidation state (Archuleta et al., 2005; Yakobi-Hancock et al., 2013)."

Archuleta C. M., DeMott, P. J., Kreidenweis, S. M.: Ice nucleation by surrogates for atmospheric mineral dust and mineral dust/sulfate particles at cirrus temperatures, Atmos. Chem. Phys., 5, 2617-2634, doi: 10.5194/acp-5-2617-2005, 2005

Yakobi-Hancock, J. D., Ladino, L. A., Abbatt, J. P. D.: Feldspar minerals as efficient deposition ice nuclei, Atmos.Chem. Phys., 13, 11175-11185, doi: 10.5194/acp-13-11175-2013, 2013

Line 64: clarify what is meant by "the efficiency of metal oxides to activate as INP depends on the type of metallic particle". Do you mean the type of metal cation?

With "type of metallic particle", we meant the type of cation as well as the oxidation state. We adjusted the sentence in accordance to this explanation.

"Their efficiency to activate as INPs depends on the type of metallic cation as well as on the oxidation state (Archuleta et al., 2005; Yakobi-Hancock et al., 2013)."

Line 83: All the references provided here seem to be for measurements of INP concentrations only. Add references specific for the mentioned identification of particle type and size.

We rearranged this part. The general description of laboratory experiments, field experiments and modeling studies was removed due to a more focused introduction, as it was suggested by the reviewers. The references for the experiments were moved to the following section. There it is clearly stated, that the references refer to concentration measurements, so no further references are needed.

"Although there is a variety of methods to determine the INP concentration in the laboratory (Hoose & Möhler, 2012; Hiranuma et al., 2015; DeMott et al., 2018; Hiranuma et al., 2019) and in the field (Wex et al., 2019; Schrod et al., 2020b; Brasseur et al., 2022; Lacher et al., 2024), only a few of them are simultaneously able to report on the chemical characteristics of individual nucleating particles."

Line 85-89: The logic is not clear in these sentences. Clarify if it is the aerosol composition, particle class, main type, or specific type that should be related to INP to improve simulations.

To combine observational data and model simulations Burrows et al. (2022) identified a key need for a closure study that combines size-resolved aerosol composition measurements, particle class dependent INP parametrizations and INP measurements. In order to evaluate the results of such a closure study in the most comprehensive way, the INP measurements ideally contain, in addition to the INP concentration, information on the size and chemistry of the ice-forming particles. However, gaining this desired set of experimental parameters from field measurements is challenging.

Such an experiment could be conducted as follows: First you have a type-specific, and best size-dependent parametrization (which don't exist in this detail yet). Then you measure the aerosol size-distribution, and best composition in order to use this data to put into your parametrization or use model data as input variables. Then you check the resulting INP concentration with INP measurements and when available you can also check the INP composition with what individual type-specific parametrizations predict.

Nevertheless, this section was removed in the revised version in order to give the introduction less of a review character.

Line 101-103: If for IR, scavenged particles cannot be distinguished from INP, there is no information on the INP.

Yes, there is a risk that collected ice crystals contain scavenged particles or that they have formed by secondary ice formation. This is, why they are referred to IR and not as INPs.

However, this discussion on INPs/ IRs was completely removed from the revised manuscript, due to a more focused introduction.

Line 103-104: Clarify how INP can be identified using a CFDC by activation of sampled non-activated aerosol.

What was meant here was that, in contrast to ice crystal collection, the entire aerosol is collected and then activated under defined conditions. We revised the sencences.

"In another approach, the particles are activated under defined conditions in an online reaction chamber (e.g., Rogers, 1988) after the collection of the total aerosol. To analyze the activated particles, it is necessary to separate the ice crystals from droplets and evaporate the ice by one of the specialized inlet systems or a droplet evaporation zone. In a second step, the separated INPs/IRs are then either analyzed in the air stream or separated and transferred to an offline analysis."

Line 106-107: specify what is meant by the "appearances of particles" and explain what observable differences can result from the evaporation process.

At this point, the physico-chemical properties of the particles are meant, especially the morphology. Since soluble/volatile components may also evaporate during the ice evaporation process, this can change the properties of the particles. This means that on a morphological level, for example, information on the original particle shape can be lost, while on a chemical level, for example, highly volatile organic coatings can no longer be detected. These sentences were also removed due to a more focused introduction. We now mention the potential changes in Section 2.6, where the individual particle analysis is described.

"The INPs have been processed in FRIDGE (multiple activation / evacuation cycles) and they were analyzed in a high vacuum under the electron beam. Therefore, the morphology of the particles may have undergone alterations. This may be especially the case for soluble / volatile components within a sample, which may evaporate during the analysis procedure."

Line 111: specify what problems online methods can run into with low INP concentrations.

Online systems may have problems to resolve low concentrations of INPs as they have typically a lower sampling flow, resulting in low INP counts in the range of background counts. Offline methods, on the other hand, can enrich the number of INPs on the sample substrates due to higher sampling flows and longer collection times.

Line 129-130: Clarify how the property influencing ice nucleation can be isolated using SEM. It would be more precise to state that the chemistry, shape and size of INP can be obtained.

It was meant in this sense. We changed it as suggested.

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

Fig.4: The perspective of the SEM image on top of the EDX spectra is confusing. Why is there a large shadow?

The image is a BSE image. The position of the particle in relation to the detector can lead to this shadowing effects.

Line 151: Schrod et al., 2017 state that: "From the present SEM analysis we cannot draw conclusions on the chemical composition and nature of INPs, which make only a 10^-3 to 10^-5 fraction of the randomly selected particles on a wafer." This was obviously a different FRIDGE-SEM-coupling, and the novelty of the method presented here should be highlighted. However, the description of chemical analysis of wafers provided in He et al., 2023 seems to already describe the current method.

With regard to the Cyprus study (Schrod et al., 2017), it should be noted that the objective was not to specifically investigate the individual INPs. Rather, the ambient aerosol on the wafer was analyzed in a random scan.

The method described by He at al. (2023) is based on our method, although the coupling of the devices (crystal detection, etc.) does not correspond exactly to our method. From the paper it is not clear, how they define the coordinates for the individual INPs from the ice crystals grown in FRIDGE (which is important to make sure that you re-find the INP and not just a particle on the wafer surface), as well as on how they define a particle unambiguous. For a paper with focus on the results, this may be sufficient, but a discussion on potential restrictions with respect to the results is missing in this paper. Our manuscript details the method, identifies strengths and weaknesses to give the reader an idea on how to interpret the results from such a coupling.

Methodology

This section resembles an operation manual, and it doesn't substantially go beyond Schrod et al., 2016 and He et al., 2023.

We can understand that the reader gets the impression of an operation manual. To lose this impression, we have restructured this section and changed the wording in some parts. In particular for the FRIDGE method already evaluated by Schrod et al. (2016), we focused more on the points that are important for the coupling procedure.

We do strongly disagree with the second point. Schrod et al. 2016 evaluates FRIDGE and He et al. 2023 essentially shows results of this method. Neither paper describes the coupling with its strengths and weaknesses as it is the case in this manuscript. For more details, see our argumentation further above.

I'm missing an explanation on how the ice is evaporated between the FRIDGE experiment and the SEM, and an analysis if IR are moved during the process.

The ice is evaporated after each measurement cycle in FRIDGE, consequently also at the end of a measurement before the wafers are transferred to the SEM. Of course, a particle drift during the FRIDGE measurement cannot be completely excluded, but measures are taken to limit the effect of a potential particle drift, if they actually appear.

"For the coupling procedure it is beneficial to stop the growth of ice before individual ice crystals grow to large sizes or coalescence, because the determination of the ice crystal center (Sect. 2.4), which is assumed to be the position of the INP, is more precise with small crystals. Additionally, this also reduces the spatial extent of potential particle drift during the ice crystal growth. By directly evaporating the ice crystals at the end of a measurement cycle with the objective of avoiding the liquid phase, the risk of possible particle drifts is also reduced."

Even if particle drift occurs during ice growth, this does not affect the results for clearly identified INPs. The particles would then no longer be in the center of the ice crystal causing only a blank position.

"A blank position may be the consequence of possible particle drift during the processing in FRIDGE (Sect. 2.3), or the result of an erroneous calculation of the ice crystal origin (Sect. 2.4)."

Line 267: quantify the coordinate uncertainties and discuss where the uncertainties come from. Based on the pixel size and the criteria of 30 pixels to identify an ice crystal location, the INP could be up to 300um away from the centre if the crystal grows as needle. It could be explained in more detail why 50um is a good value. Is it because at the investigated conditions the ice growth regime is plate like?

This uncertainty is based on the calculation of the ice crystal origin and the coordinate calibration. We added a comment on the uncertainty for the ice crystal calculation in Section 2.4. In our configuration (low pressure of near vacuum, temperatures usually -20 to -30°C), the ice crystals usually grow rather radially symmetrical, but not perfectly, hence needle-like crystals are not observed.

"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 FRDGE. Nevertheless, a potentially imperfect radial symmetry of the ice crystal growth, coupled with the restricted resolution of the FRIDGE images (20 x 20 µm), may result in an uncertainty in the calculation of the ice crystal origin. As the size of an ice crystal increases, the probability and extent of such a non-symmetrical growth also increases. The quantification of this uncertainty proved to be difficult, as it depends on the symmetry deviation present. 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."

The coordinate calibration uncertainty is mentioned in Section 2.5.1.

"Due to the limited resolution of the FRIDGE images of about 20 x 20 µm, the calibration has of course an uncertainty in the same order of magnitude."

Regarding the accuracy of the coordinate, the following is stated in the INP identification section (Section 2.5.2):

"Each ice crystal position, based on a real grown ice crystal, is inspected by SEM to identify the presence of particles. Given the uncertainties associated with the ice crystal identification process (Sect. 2.4) and the coordinate calibration (Sect. 2.5.1), it is crucial to consider not only the exact calculated coordinate but also the surrounding area. This area must take into account the aforementioned uncertainties and, at the same time, limit the probability that several particles will be observed in the scanned area. In this context, a radius of 50 µm has proven to be useful. While the previously discussed uncertainties may suggest a lager radius to be beneficial, in fact, the high substrate loading often proves to be the limiting factor."

This is also illustrated in a new Figure (Fig. 3 in the revised manuscript).

Line 276: please define refractory particles in this context.

Refractory particles are particles that are stable under the electron beam (defined in Ebert et al 2024).

Ebert, M., Weigel, R., Weinbruch, S., Schneider, L., Kandler, K., Lauterbach, S., Köllner, F., Plöger,F., Günther, G., Vogel, B., Borrmann, S.: Characterization of refractory aerosol particles collected in the tropical upper troposphere-lower stratosphere (UTLS) within the Asian tropopause aerosol layer (ATAL), Atmos. Chem. Phys., 24, 4771-4788, doi: 10.5194/acp-24-4771-2024, 2024

In the original version of the manuscript, we used the term refractory for the particles that can be analyzed with our method. According to the definition by Ebert et al. (2024), this term is not entirely correct for our particles, as we can also analyze particles that are partly unstable under the electron beam. In the new version of the manuscript, the term refractory is no longer used.

Line 277: it is unclear what is meant by "with respect to the analysed particles" here.

We agree that the phrasing in this line is somewhat unclear. We wanted to say that the particles that can be analyzed with this method are not affected by the loss of volatile particles. Due to the restructuring this part was removed in this section. It is now part of the single particle analysis (Sect. 2.6).

"At this point, it should also be noted that our findings revealed an absence of small volatile compounds on the wafers in the EM, which are typically present in larger numbers in the total aerosol. Presumably, there is a loss of these components during sampling collection or processing. However, as these volatile particles are not

known to be efficient INPs in the considered temperature range (Murray & Liu, 2022), it can be assumed that their absence does not significantly affect the results."

Line 279: Explain why adjusting the scanning radius optimizes the analysis. It could be quantified on a lightly loaded wafer what the distance of INP from the coordinate usually is to exclude particles outside the range.

Our chosen radius of 50 µm represents a compromise. On the one hand, as already discussed above, possible uncertainties in the coordinate calculation and calibration in the SEM must be taken into account. On the other hand, the radius must not be too large, as otherwise the chance of finding several particles in the selected radius increases. The number of particles in the corresponding scanning radius depends strongly on the total wafer load and often represents the limit for the selected radius. If the wafers are heavily loaded, hardly any particles can be unambiguously identified. For lightly loaded wafers, expanding the scanning radius can provide an opportunity to assign more particles to ice crystals and thus increase the yield.

Line 285: Explain how it can be known if a feature is relevant for ice formation.

This should just be a list of particle features that can be analyzed with the SEM method which may be important for its ice nucleation ability. We do not say that we know explicitly what exactly make a particular particle form ice from our analysis. We changed the wording to physico-chemical properties instead of mixing state and distinct morphological patterns on the particle surface.

"The comprehensive single particle analysis (Sect. 2.6) enables the identification of physico-chemical properties that may be pertinent to ice formation."

Line 299-302: Describe the stage where ice is evaporated before the SEM analysis.

The ice is evaporated after each measurement cycle in FRIDGE. So, all the ice is gone before the wafer is taken to the SEM.

"By directly evaporating the ice crystals at the end of a measurement cycle with the objective of avoiding the liquid phase, the risk of possible particle drifts is also reduced."

2.6 Chemical classification: Fig. S1 could be shown here and referred to, to guide the reader and help to follow the descriptions.

The Figure was added to the main paper.

Line 307: Again, how can be identified if a certain surface property is relevant for ice nucleation?

In the current setup we cannot investigate this in detail, that's true. But in general, it's possible to analyze the surface properties of the identified INPs. Even if we cannot define the exact location where ice growth started, we can analyze the INP surface for structures that can promote ice formation.

Line 315: Explain, based on what information the classification scheme is modified. Does this make the scheme subjective?

The classification scheme is essentially based on chemistry and in some cases also on morphology. The mentioned modification does not refer to the criteria of the individual particle classes, but rather to the grouping of the particle classes based on different abundances at different locations. We added and modified the sentences to clarify this.

"In the following section we define a classification scheme, which is mainly based on elemental composition (Fig. 5) and in some cases on the morphology of particles (Fig. 7). … Based on the research question or the occurrence of specific particle classes at the sampling site, the classification scheme may be adapted."

Fig. 3: x-, y-axis scale are too small to read. Also, axis labels should be added.

The scales are increased and the missing labels are added. The y-axis of the EDX-spectra are not as meaningful, as they show only the counts, which are dependent on the EDX sampling time and the current of the electron beam.

Case Study

Clarify if sampling was conducted downstream of an inlet or in the open.

We added a sentence to clarify this.

"FRIDGE sampling was conducted downstream of the GAW total inlet (Lacher et al., 2018)."

Line 391: Define cINP as INP concentration.

We removed the cINP abbreviation due to rare occurrence in the script.

Fig.4: increase the contrast of the figure. Corresponding sample numbers are not shown. In the caption, do you mean adapted from Weber (2019) instead of modified according to Weber (2019)? Specify that the 5-day average is a running average.

*Fig. 4 (now Fig.8) has been adjusted: Corresponding sample numbers were added.
In the caption we changed "modified according to Weber (2019)" to "adapted from Weber (2019)" and it is now specified in the caption that is a 5-day running average.*

Line 402: Provide a reference for Saharan dust being active below -20°C.

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., Leisner, T.: A Particle-Surface-Area-Based Parametrization of Immersion Freezing on Desert Dust Particles, J. Atmos. Sci, 69 (10), 3077-3092, doi: 10.1175/JAS-D-11_0249.1, 2012

Murray, B. J., O'Sullivan, D., Atkinson, J. D., Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, Chem. Soc. Rev., 41, 6519-6554, doi: 10.1039/c2cs35200a, 2012

The references were added to the manuscript.

Line 402: From Fig. 4 it is not clear when the 14 samples were taken. 31 cINP on 11 days are marked with triangles.

We added the corresponding sample numbers to Fig.4 (now Fig. 8). Now it should be clear, on which days the 14 samples were collected. For each sample, the INP concentration for -20°C, -25°C and -30°C was marked with a triangle, that's why there are more than 14 triangles.

Fig. 5: What's the point of this figure? As shown in Fig. 2 and explained in Sec. 2.4. the edge region ice crystals were excluded for SEM. There should therefore be clearly more ice crystals detected by FRIDGE than SEM positions.

This Figure and the related analysis are moved to the section 2.4 in the method part, as suggested by one reviewer. We have therefore moved Figure 2 of the original script to the supplement.

The point of this figure is to show that two separate ice crystal identification algorithms (FRIDGE and the one discussed in section 2.4) come to similar results, which serves to validate the efficacy of our software-based approach to identify the ice crystals from the from the FRIDGE images. Note, that the edges are also excluded in FRIDGE, the counting areas are the same for both methods.

In the caption, mention that the 1:1 line is shown in red.

Changed as requested.

Line 420: Clarify why the algorithm is inconsistent with excluding the temperature sensor area.

Based on the standard wafer positioning in FRIDGE, the counting algorithm has a given area in which the ice crystals are detected from the FRIDGE images. Normally, the area around the temperature sensor is excluded from the analysis. However, in some cases it may happen, that the wafer is not correctly positioned is FRIDGE, which can lead to the temperature sensor being in the counting area of the algorithm. The counting algorithm is not able to identify this by itself, and counts all differences in brightness around the sensor as ice crystals.

Further improvements in the calculation of individual ice crystal positions can be achieved by specifically avoiding the sources of error previously mentioned.

This part was removed in the revised manuscript as it only represents a special case and should be avoided according to the description in FRIDGE.

Line 437: Explain why a meteorological interpretation is not feasible for the current manuscript. Chapter 6 in the thesis of Weber 2019 contains a meteorological interpretation of the JFJ results and Fig.6.18 therein shows a comparison during and outside SDE.

Here we wanted to say that such a detailed interpretation goes beyond the scope of this manuscript, submitted for AMT. Moreover, the number on identified particles is low. This part was rewritten.

Nevertheless, we decided to show the subdivision into SDE and non-SDE, as it shows that the method allows statements to be made about INP-relevant trends due to its high accuracy of identification despite low INP numbers.

Fig.6: Fig. S2 implies that the composition of the INP population active at -30°C can substantially vary from day to day. E.g., looking at W2, W7, W11 and W34 where a similar number of particles were analysed, the abundance of components is never similar. Please analyse and discuss the implications on sampling statistics and what the total chemical composition in Fig.6 represents.

Of course, the chemical composition can vary from day to day. Regarding the main particle classes, from the previously mentioned samples, only W7 looks significantly different, as it has very few mineral particles. Overall, the composition is rather similar.

The requested evaluation was added to the manuscript as follows.

Section 3.2: „Although the number of identified INPs appears comparatively low for a campaign period of five weeks, these INPs were identified with a high degree of reliability (Sect. 2.5.2). The small number of particles identified bears the risk that individual, time-limited variations occurring randomly during the sampling periods may influence the resulting total composition toa certain degree. It should therefore be noted that the results presented below do not comprehensively reflect the main composition of the INPs over the entire campaign period. Nevertheless, it can be shown that the method provides valid results for the main groups of INPs (see confidence intervals for Fig. 9 in the supplement (Tab. S2)).“

Section 3.3: "Due to the limited number of identified INPs per sample (Fig. S3), mapping daily fluctuations is not possible for this campaign. Figure 9a) provides the chemical composition for all INPs sampled over the entire campaign period, within the restrictions mentioned in Sect. 3.2."

In the caption, what artifact is excluded?

A particle with traces from gold is excluded, as gold may originate from our gold wires inside the EAC. This now clarified in the text.

"One particle with attached gold traces was classified as an artifact and therefore excluded from further discussions."

Line 487-488: It is mentioned that a comparison of abundance is not possible. Clarify the purpose of doing a comparison if the abundance, which is the main result, cannot be compared.

We stated that the particle classes they found for their IRs are similar to our INP particle classes, which represents an initial comparison. However, since the activation conditions have a substantial influence on the activation of an INP, it is generally difficult to compare chemical compositions of INPs that have been activated under different conditions. Therefore, we just mentioned, that is not possible to compare the distinct abundances for different particle classes, because of different activation conditions.

However, the discussion part has been restructured. We are now focusing more on supporting our data with results from other studies rather than on discussing differences and mentioning points which we cannot compare.

Line 505: Clarify how the agreement can be considered good if only a rough comparison can be made. The comparison suffers from the previous mentioned differences in activation temperature and overall technique, making a direct comparison questionable.

We agree and refer to our previous response.

Line 537: Specify the role of volatility.

Typically, the size distribution of an atmospheric aerosol is shifted towards smaller diameters, compared to our INP size distribution. We assume, this is the case for two reasons: On the one hand, particles with a diameter from 0.5 µm are known to be more ice active in the considered temperature regime, on the other hand, we have noticed an absence of small volatile compounds on the wafer in the SEM.

The particle loss is mentioned in Section 2.6:

"At this point, it should also be noted that our findings revealed an absence of small volatile compounds on the wafers in the EM, which are typically present in larger numbers in the total aerosol. Presumably, there is a loss of these components during sampling collection or processing. However, as these volatile particles are not known to be efficient INPs in the considered temperature range (Murray & Liu, 2022), it can be assumed that their absence does not significantly affect the results."

Fig. 7: Clarify if analysed particles activated at all of the listed RH's or at least one.

In FRIDGE all samples were activated at a set of 3 temperatures (-20°C, -25°C and -30°C) and a minimum of 4 relative humidities (95% / 97% / 99% / 101%). Since measurements at higher humidities typically show a larger number of ice crystals, we have chosen the measurements cycles at RH=99% and RH=101% for the coupling procedure. RH = 95 / 97% was chosen for one sample due to cluster formation at higher RH. We added the information to the corresponding figure captions.

Line 549: This is a misunderstanding. DeMott et al. 2010 found that the concentration of INP correlates to the concentration of particles >500nm. Not that they are >500nm.

This is basically correct, but from the results of DeMott et al. (2010) it is very likely that INPs are often larger than 500 nm, surely not all INPs are larger than 500 nm. The larger the particle the higher is the probability of active sites on the surface.

"This agrees to well-established findings in the literature substantiating that most particles that act as effective ice nuclei are above a size of 500 nm (DeMott et al., 2010)."

Line 569: Quantify the statistical uncertainty."

We have quantified the confidence intervals for the chemical composition given in Fig. 9 (revised version). The values are shown in the supplement (Tab. S2). In addition, all particles with a d_{pa} larger that 6 µm have been summed up, due to their low abundance.

Line 573: Looking at Fig.7 in Lacher et al., 2021 the second maximum in the OPC data appears between 0.5.1um. Clarify how the IR OPC data is compared to the current results.

We mean the broad maximum from 2-5 µm (OPS) and between 2-3 µm for the Sky-OPC. This indicates an enrichment of particles with larger diameters, supporting our theory of an enrichment of larger particles in the ice active fraction.

Line 574: Is a comparison just difficult or not possible?

We now agree that a meaningful comparison is probably not possible due to the different sizes of the identified particles. In this paragraph (comparison of the size-resolved chemical composition), we have decided to refrain from a comparison with Lacher et al. (2021) and limit the comparison to Worringen et al. (2015).

Line 586: Explain how the size of ice crystals is linked to the size of INP/IR.

The description of Ice-CVI by Mertes et al. 2007 states that only ice crystals between 3 and 20 µm are extracted, as the probability of scavenging is low for them. If an unactivated INP already has a size close to or above 20 *µm, the resulting ice crystal will probably be larger than 20 µm and will therefore not be extracted. So consequently, there is a size limitation for big particles.*

This sentence was removed for the revised manuscript, because it does not fit into the new text.

Line 589: Better references for the importance of immersion freezing are Ansmann et al. 2009 or Westbrook and Illingworth, 2011.

Changed to Ansmann et al. (2009). Additionally, Murry et al. 2012 was also added as a reference.

Murray, B. J., O'Sullivan, D., Atkinson, J. D., Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, Chem. Soc. Rev., 41, 6519-6554, doi: 10.1039/c2cs35200a, 2012

Line 591: It needs to be pointed out clearly what part of the presented method is novel.

We now state in the conclusions as well as in the introduction, that the method was already used in campaigns, and that the discussion from a methodological perspective in new. For more details, we refer to the previous comments and our answers to the other reviewers.

Section 1: "The FRIDGE-SEM-coupling technique has been used for several campaigns in recent years, providing valuable results (Schrod et al., 2017; Schrod et al., 2020b; Weber, 2019; He et al., 2023). Details of the FRIDGE method were described by Schrod et al. (2016)."

Section 4: "A method for analyzing the concentration and individual physico-chemical properties of ambient INPs, which has been used in several campaigns (Schrod et al., 2020b; He et al., 2023), is discussed here from a methodological perspective. The method benefits from the coupling of two instruments already used for the analysis of INPs and IRs: the static diffusion chamber FRIDGE and the SEM. As the individual methods are already known, the focus here was on a description of the coupling and the associated advantages and uncertainties, as well as the resulting potential of the method."

Line 594: The analysis of morphology and surface properties has not been demonstrated in this work.

The morphology is now explicitly mentioned in chapter 2.6 ("individual particle analysis") and an example for a mixed particle is also provided. The possibility to identify surface properties is given by the method, but not discussed in the case study. But since the focus of this paper is to highlight the possibilities of the method, we find it important to point out that this is generally possible.

Line 606: Specify what improvements are necessary.

Improvements are generally necessary in the wafer cleaning process, in improving the uncertainties during the coordinate calculation for the ice crystal origin, and in increasing the identification rates.

The knowledge about the background counts for clean wafers can be improved, for example, by checking every cleaned wafer and not just random samples from a set of cleaned wafers.
The uncertainties in crystal detection could be reduced by the following. The shortest possible measuring time in FRIDGE enables a more precise determination of the crystal origin and minimizes the risk of particle drift. Another improvement could be a camera with a higher resolution in the FRIDGE setup. This could increase the resolution of the FRIDGE images, which would lead to a more precise recovery of the calibration point on the SEM.
A systematic, automated analysis of the INPs could possibly also reduce the working time and thus open up the possibility of a larger number of samples.

Line 625-627: It has not been demonstrated in this work that meaningful structural information can be obtained, and it is unclear how information on the relevance of a property for ice nucleation can be gained with this method.

We have revised the sentence.

"The detailed information on physico-chemical particle properties that can be obtained from SEM can be a valuable addition to pure INP counting methods for gaining information on the relevance of particle properties

to ice nucleation efficiencies and could help to bridge the knowledge gap towards INP aerosol-type-specific parametrizations that could be used in modeling studies (Burrows et al., 2022).”

Line 631-632: Clarify what element of the method need adaptation. It can be assumed that only the EAC would be flown, and the method of analysis remains the same. Also, is the EAC not already usable in an aircraft setting?

A prototype was used in 2011 in an aircraft campaign, but the samplers had to be rebuilt in terms of material and adjustable parameters due to the different environmental conditions in the UT/LS. For example, it is assumed that the HV has to be reduced to avoid flash overs in regions with lower pressure.

But also, the FRIDGE measurement procedure has to be adapted. FRIDGE has to be operated at colder temperatures to get closer to the conditions in the UT/LS region for example. Therefor the wafer cleaning has to be improved, as the background counts typically rise sharply for measurements at -35°C.

Technical corrections:

Line 48: I can’t find information about hydrogen-bridging functional groups in Kanji et al., 2008. Double check the reference.

This is a mistake on our part. The reference Kanji et al. (2008) was incorrectly placed as a reference for hydrogen-bridging. It has been moved accordingly as a reference for coating.

“In addition to the prevailing environmental conditions i.e., temperature and humidity, the potential for an INP to become activated is dependent upon individual particle properties (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).”

Line 51: Replace Hoose & Möhler, 2012 with a more specific reference about the influence of particle size on ice nucleation.

The reference has been changed to: Welti et al. (2009)

Welti, A., Lüönd, F., Stetzer, O., Lohmann, U.: Influence of particle size on the ice nucleating ability of mineral dusts, Atmos. Chem. Phys., 9, 6705-6715, doi: 10.5194/acp-9-6705-2009, 2009

Line 52: Marcolli 2017 would be a more specific reference for pre-activation than Abdelmonem et al., 2020.

Changed as requested.

Line 174: I can only find information on the EAC in the Supplement of DeMott et al., 2018 and it is not clear how the EAC was modified compared to the description in Klein et al., 2010.

The EAC is described in detail in Klein et al., 2010 and in Schrod et al., 2016. Our intention here was to state that different versions have been used but not that they are described in detail in the given literature. We rearranged the section:

“Aerosol is precipitated onto the substrates using an Electrostatic Aerosol Collector (EAC) (Klein et al. 2010). Several EACs have been deployed for the use in the laboratory (DeMott et al., 2018), in field campaigns (DeMott et al., 2024), for measurements with unmanned aerial vehicles (Schrod et al., 2017), and for long-term observations at research stations (Schrod et al., 2020b). The most recent version, which was also used in the case study (Sect. 3), PEAC7, is a programmable EAC (Schrod et al., 2016) designed for semi-automated operation for one week of daily sampling.”

Line 175: Lacher et al., 2024 report FRIDGE measurements using filter samples, the EAC seems not to have been used.

Yes, that is a mistake. The EAC was used but the corresponding measurements were not included in the fonal study. We removed the reference.

Line 270: "two effects" instead of "to effects"

This section has been rewritten. The wording no longer occurs in the new version.

Line 496: superfluous)

The complete case study discussion was restructured. The sentence was deleted.

References:

Abdelmonem, A., Ratnayake, S., Toner, J. D., Lützenkirchen, J.: Cloud history can change water-ice-surface interactions of oxide mineral aerosols: a case study on silica, Atmos. Chem. Phys., 20, 1075-1087, doi:10.5194/acp-20-1075-2020, 2020

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Author response to Reviewer 3

First of all, we would like to thank the reviewer for reading and evaluating our manuscript. The comments helped us to improve the manuscript and strengthen the desired focus on the methodological part.

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.

In this manuscript, the authors present an offline method that combines an ice nucleation counter, the FRankfurt Ice nucleation Deposition freezinG Experiment (FRIDGE), with Scanning Electron Microscopy (SEM) to analyze the chemical composition, size, and morphology of Ice Nucleating Particles (INPs) collected from ambient air. The authors begin by providing an overview of the methodology, followed by a case study demonstrating its application to ambient aerosols collected during the 2017 CLACE/INUIT campaign at the Jungfraujoch station.
The methodological section appears unfinished, as it fails to demonstrate all the potential features the authors claim the method can analyze (e.g., morphology, pores). Additionally, the lack of standards in this section raises concerns about evaluating the performance of the technique and estimating statistical error, particularly for size measurements and the coupling of particle size with ice nucleation efficiency.

The methodological part of the manuscript has been improved. Uncertainties (e.g., for the identification of ice crystal origins (Sect. 2.4 in the revised manuscript) and the coordinate calibration for SEM (Sect. 2.5.1 in the revised manuscript)) are now defined and discussed more clearly. Please note that the pure FRIDGE method including statistics has already been evaluated by Schrod et al. (2016).

Sect. 2.4: "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 FRDGE. Nevertheless, a potentially imperfect radial symmetry of the ice crystal growth, coupled with the restricted resolution of the FRIDGE images (20 x 20 µm), may result in an uncertainty in the calculation of the ice crystal origin. As the size of an ice crystal increases, the probability and extent of such a non-symmetrical growth also increases. The quantification of this uncertainty proved to be difficult, as it depends on the symmetry deviation present. 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."

Sect 2.5.1: "Due to the limited resolution of the FRIDGE images of about 20 x 20 µm, the calibration has of course an uncertainty in the same order of magnitude."

The method evaluation from the case study was also included in the methodological section. The comparison between the different ice crystal counting methods (formerly Fig. 5) was moved to the method section (Section 2.4 "identification of ice crystal positions") and replaces the graphical representation of coordinate determination (formerly Fig. 2), which has been moved to the supplement. The discussion on the identification rates was included in section 2.5.2 (now called "INP identification").

Sect. 2.5.2: "The number of INPs that can be unambiguously attributed to an ice crystal origin is significantly influenced by the total wafer loading, which is determined by the sampling parameters (e.g., flow rate, sampling time, deposition efficiency) in combination with the aerosol concentration present. However, 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. This ratio is variable and usually unknown prior to measurement. As a result, the amount of atmospheric aerosol and the proportion of INPs deposited on a wafer are highly variable. This variability is also seen in the identification rates, which is why it would be misleading to give an average identification rate for the method presented. However, a specific identification rate for the case study conducted at the high-altitude research station Jungfraujoch (Sect. 3) can be given here as a guideline. The average INP identification rate was calculated to be 30% (ranging from 13% to 50%). Furthermore, the study identified the presence of multiple particles at 45% of the locations (ranging from 7% to 81%), while the remaining 25% (ranging from 2% to 66%) were found to be blank positions."

Section 2.6 (now called "individual particle analysis") has been generalized, to show the potential of the coupling method. The morphology is now mentioned in Sec. 2.6 as it can be used to classify INPs. We included *also a BSE picture showing a particle with different chemical compositions on its surface (Fig. 4 in the revised manuscript), to illustrate that it is also possible to see the element distribution on the particle surface.*

"The analysis by SEM and EDX is an efficient method for characterizing INPs in detail, as it provides information on elemental composition and distribution as well as on morphology and surface properties. The morphological information can be used for source apportionment (e.g., biological particles, soot, spherical particles from high temperature processes). With this detailed information, it is possible, for example, to determine the mixing state of a particle (see Fig. 3 and Fig. 4)."

The size of the particles can be determined with high precision due to the high resolution of the electron microscope, in contrast to the FRIDGE camera, where the resolution is limited.

For more details we refer also to our following responses and our comments to Review 1 and 2.

The case study lacks sufficient statistical analysis and fails to provide a clear connection to other parameters measured during the campaign (e.g., aerosol size distribution and number concentration).

The focus of the manuscript is the methodological description and discussion of the FRIDGE-SEM coupling. Therefore, the case study Section was shortened. Nevertheless, for statistical purposes, we have calculated the confidence intervals (Supplement Fig. S2) for the chemical composition shown in Fig. 9 (revised manuscript). Additionally, we have connected our INPs size measurement to the aerosol size distribution from the CLACE INUIT campaign provided by Weber (2019). For more details see our comment on Fig, 7 below.

It appears the authors did not clearly decide whether to (A) describe and evaluate the method in detail, suitable for AMT journal or (B) focus on the CLACE/INUIT campaign with further analysis. As a result, the manuscript presents two incomplete studies that are not well connected.

We can see that a reader may get this impression. In fact, the decision between ACP and AMT was under discussion for a long time. In the end, we decided on AMT, but it seem that the manuscript was not sufficiently adapted to this decision.
For the revised version, we have placed a clear focus on the methodological discussion and shortened the case study.

Specific comments

- A clearer explanation is needed as to why deposition and condensation freezing modes are grouped together as the two primary ice nucleation modes. Specifically, the cycles with FRIDGE include measurements taken below water saturation at RH=100%, which would typically prevent condensation freezing. Is this grouping due to uncertainty in the RH measurements, which could allow for RH to exceed 100%?

Apart from an uncertainty in the RH measurement, the two nucleation modes are mainly combined, as we measure both below RH=100% water saturation and specifically beyond RH=100% (95% / 97% / 99% / 101%). While INPs are activated solely by deposition freezing at RH distinctly below 100%, condensation freezing also takes place around/above 100%.
As more INPs are usually activated at higher RH, the series of measurements around RH=100% are better suited to the coupling method.

- The authors argue that volatile compounds are not detected and that these compounds are generally not known to be efficient INPs. Is there any estimation of which type of volatile material is lost, a lower estimation of vapor pressure? How does this affect SEM measurements, notably for carbon? Could there be potential effect of freezing point depression, such as the competition for adsorption on active ice nucleating sites between water and other volatile compounds?

We work under near-vacuum conditions in both the FRIDGE and the ESEM. Before starting the measurement, a vacuum is created in the FRIDGE chamber ($p<<0.1$ mbar). After each measurement cycle, the chamber is evacuated again until the chamber pressure from before the measurement is restored. This is to ensure that all water vapor that has been added to the chamber during the measurement, and thus also the ice crystals, is removed again before starting the next measurement cycle. So typically, the chamber is evacuated 13 times for one sample. In the SEM, the samples are analyzed in a high vacuum at approx. 10^{-6} mbar. All compounds, which are stable under the vacuum conditions can be analyzed with SEM/EDX.

A second limitation is the stability under the electron beam. All particles which are not stable under the electron bombardment can possibly be seen, but no chemical classification is possible as they evaporate during the spot analysis. In SEM, HVOCs and (ammonium)nitrates are particularly problematic, whereas sulfates, nitrates and especially carbon-rich particles as an important INP compound can be analyzed by SEM/EDX. As a consequence, the limitation for the analysis of volatile particles is not given by SEM.

Nevertheless, we observe that the dominant secondary atmospheric particles of the atmosphere are not visible on the wafers in the EM. It is therefore reasonable to suspect that these particles are lost during the sampling collection or processing. In the manuscript we say the following:

"At this point, it should also be noted that our findings revealed an absence of small volatile compounds on the wafers in the EM, which are typically present in larger numbers in the total aerosol. Presumably, there is a loss of these components during sampling collection or processing. However, as these volatile particles are not known to be efficient INPs in the considered temperature range (Murray & Liu, 2022), it can be assumed that their absence does not significantly affect the results."

- The case study part focuses primarily on chemical composition and size analysis, but the title of the paper also mentions morphology. The conclusion, line 594, states, " This coupling allows for detailed analysis of various INP properties, such as chemical composition, mixing state, size, morphology, and surface properties like cracks or pores." Yet, no information is provided regarding the mixing state, morphology or surface properties.

It is true that the case study was primarily focused on the chemistry and size of INPs.
Nevertheless, the detailed single particle analysis by SEM and EDX provides, in addition to the chemical composition, information on the surface structure (secondary electrons) and the element distribution (backscattered electrons). Even if it is not possible to define the exact origin of the ice growth on the particle surface, with the current setup, the method can generally be used to investigate the mixing state, morphology and surface of the identified INPs.
The potential of the method is described in more detail in the new version of the manuscript in Section 2.6. There we mention, that the morphology can be used to identify certain particle classes (soot, biological particles) and sometimes to determine the origin of particles (e.g., irregular geogenic minerals vs. fly ash). We have also added an example of a mixed particle which, in contrast to the mixed particle in Fig. 3 of the old manuscript, allows clear differentiation of the regions with different chemistry.

- Figure 3: The x and y axis are not readable. Only few EDX spectra are provided in figure 3 for the case study analysis. How can we evaluate the reliability of the chemical composition on all particles?

The labeling of the x-axis has been adjusted. The y-axis provides information on the number of counts which depend on the parameters set during the EDX analysis. The exact numbers are of no further importance for the evaluation carried out here, the peak intensities are clearly visible.
The entire description of the particle classes was isolated from the case study and generalized. This section now highlights the potential and discusses the limitations of this method for analyzing INPs. We provided example EDX spectra in Fig. 7 (revised manuscript) to illustrate the classification of INP types. We think it would be overly excessive and uninformative to present those for all particles.

- Figure 4: How is the 5-day average calculated and plotted? Why are there no error bars? What is the background level? How many measurements were performed per wafer, 1 only? Schrod, J. et al. (2016) provided statistical analysis for FRIDGE.

The 5-day average is a running average which is calculated for each day from the two previous, the current and the two following daily values.

For a detailed discussion and analysis of the INP concentrations at the JFJ, error bars would be essential, we agree with the reviewer. However, such a discussion is not within the scope of this paper. At this point, we show the Figure to give the reader an overview and to illustrate why we focus on the activated INPs at -30°C for the analysis. In this case, we see no need to discuss uncertainties in the INP measurement in depth. This FRIDGE method was evaluated by Schrod et al. (2016).

As you have already mentioned, Schrod et al. (2016) provides a statistical analysis of the FRIDGE method. A value of 20% is given for the statistical fluctuation of the determined ice crystal number. A statistical fluctuation *of the same order of magnitude can be assumed for the concentrations shown here, which is based on 1 measurement. This is now also described in the manuscript.*

"The concentration for each sample is calculated on the basis of one measurement. The relative error of the counting uncertainty for individual measurements is 20% (Schrod et al., 2016), so the error of the concentrations given here is also in this range."

Typically, even cleaned wafers show low ice formation activity at -30°C. As already mentioned in chapter 2.1, this is typically around 10 counts per wafer. These counts are normally subtracted from the number of ice crystals before the concentration is calculated. The concentrations are therefore usually already background corrected. For the campaign at the JFJ, particular emphasis was paid to the cleanliness of the wafers, as we expected very low INP concentrations in advance. The background counts for -30°C were less than 3 counts per wafer. This results in a maximum background concentration of 0.03 L^{-1} for a collection volume of 100 L. As the collection volumes were generally larger than 100 L, the value decreases accordingly.

- Figure 5: What is the red curve, is this the ideal version with 1:1 ratio?

Yes, the red line shows the 1:1 ratio. A description was added to the figure caption.

- Figure 6: Why is n=199, shouldn't it be 200? Are the same particles active at all RH values (95%, 97%, 99%, 101%)? The percentages should be linked to the previous figure, as you mention the total chemical INP composition, but only 15% of all INPs are considered. Additionally, there are no error bars. Soot accounts for 1% of the total INP, does this fall within the uncertainty range?

One artifact is excluded from further analysis, that's why it is only 199. This is now clearly stated in the text.

"One particle with attached gold traces was classified as an artifact and therefore excluded from further discussions."

In FRIDGE all samples were activated at a set of 3 temperatures (-20°C, -25°C and -30°C) and a minimum of 4 relative humidities (95% / 97% / 99% / 101%). Since measurements at higher humidities typically show a larger number of ice crystals, we have chosen the measurements cycles at RH=99% and RH=101% for the coupling procedure. RH = 95 / 97% was chosen for one sample due to cluster formation at higher RH. We added the information to the corresponding figure captions.

In consideration of the representativeness of our findings, we have included the following additional information.

"Although the number of identified INPs appears comparatively low for a campaign period of five weeks, these INPs were identified with a high degree of reliability (Sect. 2.5.2). The small number of particles identified bears the risk that individual, time-limited variations occurring randomly during the sampling periods may influence the resulting total composition to a certain degree. It should therefore be noted that the data presented below may not comprehensively reflect the main composition of the INPs over the entire campaign period. Nevertheless, it can be shown that the method provides valid results for the main groups of INPs (see confidence intervals for Fig. 9 in the supplement (Tab. S2))."

For the chemical composition (Fig. 9 in the revised manuscript) we have calculated the 95% confidence intervals and provide these in the supplement (Tab. S2). Of course, for the minor particle classes the differences in the abundance are within the uncertainty range, but for the major components it is possible to derive type-specific conclusions. The discussion of the results was shortened and more generalized.

- Figure 7 and discussion: No details comparison is made between initial size distribution of particles and size of INP, which do not enable correct estimation how size affect ice nucleation. In lines 556-566, the range referred here may be biased by simply higher initial concentrations. You need to better connect this part to size distribution measurements in lines 571-584, with statistical analysis.

Our INP size distribution is now compared to the particle size distribution over the whole campaign period provided by Weber (2019). The comparison with the size distribution of the total aerosol during the same campaign period shows that the maximum of our size distribution is shifted to larger diameters compared to the total aerosol. As we laid more emphasis on the methodological part of the manuscript, we chose to describe the likely enrichment of larger particles an INPs based on the size distributions presented in Weber (2019) in a more qualitative manner.

"In comparison to the total aerosol size distribution from the whole campaign period (Weber, 2019), the maximum of the INP size distribution is significantly shifted towards larger diameters. We hypothesize that, in addition to the primary suitability of larger particles as ice nuclei, the absence of the small volatile aerosol components (nitrates, sulfates, and volatile organics) may play a role here (see Sect. 2.6)."

The comparison to Lacher et al. (2021) and Worringen et al. (2015) has been rewritten and divided according to a comparison of the INP size and a comparison of the size-resolved chemical composition.

"Lacher et al. (2021) and Worringen et al. (2015) provide size distributions for INPs/IRs measured with different techniques at the high-altitude research station JFJ up to a size of 3 µm and 5 µm, respectively. In both studies, the highest concentration was found for IRs smaller than 0.5 µm, but the broad maximum (diameters between 1.3 µm and 5 µm) from Lacher et al. (2021) agrees reasonably well to our findings. The same is the case for particles collected with the Ice Selective Inlet by Worringen et al. (2015), which also showed a secondary maximum at 1 - 1.5 µm. The shift towards larger particle diameters in our results in comparison to the maxima from Lacher et al. (2021) and Worringen et al. (2015) may be caused by the differences in sampling and ice activation. INPs in FRIDGE are activated through deposition nucleation / condensation freezing under defined conditions, while the IRs collected from ambient air are activated under natural and even more complex conditions, including the potentially more important immersion freezing mode (Ansmann et al., 2009; Murray et al., 2012).

The comparison of such INP size distributions with chemical information from different methods is difficult, since in addition to the influencing factors discussed in Sect. 3.3, a possible size selection or limitation of the sampling process, and different techniques of particle sizing may also play a role. Nevertheless, the results for our main groups are in reasonable agreement with the results from Worringen et al. (2015). In our results, both the metal oxides and the few soot particles were observed at very small diameters, which is comparable to carbonaceous particles/soot and metal oxides predominantly detected in the submicron range by Worringen et al. (2015). Terrigenous particles, including silicates and Ca-rich particles, were primarily found in the larger size ranges, while our mineral components were distributed over all size ranges, with silicates domination for particles from $d_{pa} > 0.5$ *µm. In contrast to Worringen et al. (2015), our C-rich particles were present over the entire size range. The reason for this is possibly that our classification scheme assigned the larger potentially biological particles as C-rich."*

Technical corrections

- Line 350 "natural mineral durst" correct to dust

The typo has been corrected.

-Line 401: "This was not the case for -20°C, because Saharan primarily activate as temperatures below -20°C." reference?

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., Leisner, T.: A Particle-Surface-Area-Based Parametrization of Immersion Freezing on Desert Dust Particles, J. Atmos. Sci, 69 (10), 3077-3092, doi: 10.1175/JAS-D-11_0249.1, 2012

Murray, B. J., O'Sullivan, D., Atkinson, J. D., Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, Chem. Soc. Rev., 41, 6519-6554, doi: 10.1039/c2cs35200a, 2012

The references were added to the manuscript.