Summary of Author Responses to Reviewers

Reply to Reviewer 1

Author Response

We are grateful to the reviewer for their comments on the manuscript, which have helped to improve it.

We have corrected some problems with the original Figure 11. Also, we have explained our isothermal formulation (now Eq (2)) in terms of a new Eq (1) expressed in terms of the number of unfrozen drops remaining.

Point-by-Point Comments

Reviewer: General comment: The revised manuscript is clearly improved compared with the initial submission. The authors have taken the concerns of the anonymous reviewers seriously and modified the manuscript accordingly. The writing of the manuscript has been improved and imprecise formulations have been reformulated more clearly. The reasoning of the authors can now be followed throughout the mnuscript and the aims and purpose of the study have become clearer. Specifically, the following improvements have been made:

- The relevant literature is discussed more fully in the revised manuscript.

- A detailed discussion of the composition of the different aerosol classes has been added. Although the physicochemical characterization given in the revised Table 1 exhibits some gaps, it helps to better classify and compare the different samples. It is more clearly discussed to what extent the analyzed sample types are able to represent the proposed aerosol classes.

- Statistical tests have been added to analyze whether the samples exhibit significantly different freezing spectra.

- The addition of a pure water-freezing curve helps to judge that there is no relevant influence of the background signal on the freezing spectra of the samples.

- A discussion of the origin of time dependence was added and it was concluded that the time dependence is mostly stochastic.

- The empirical approach to treat the time dependence is now explained better. An appendix with a table containing the definition of the mathematical symbols has been added that helps to understand the equations.

- The conclusions have been extended. Most importantly it is explicitly stated that the stochastic component of immersion freezing is minor compared with the temperature dependence of INP freezing.

This is an important finding of this study, in view of a purely stochastic representation of immersion freezing that has been proposed in a recent study (Knopf et al., 2020).

Response: We are glad to see the reviewer appreciates our modifications.

Reviewer: Yet, the valuable input from Gabor Vali has only partially been considered in the manuscript revisions. The suggestion to show the results in terms of the freezing rate, i.e. fraction of unfrozen droplets that freeze per time, has been taken up in Fig. 11 by adding a panel (b) displaying the freezing rates of the samples. Unfortunately, throughout the rest of the manuscript, the authors stuck to an analysis in terms of their fractional freezing rate, which has much less physical meaning. Yet, even if the authors do not want to convert their fractional freezing rate to the real freezing rate, the revised manuscript can be published after the following minor revisions:

Response: Yes and no.

In fact, our isothermal freezing formulation, now Eq (2), is derived from an assumption that the unfrozen fraction decays quasi-exponentially (with a relaxation time that depends on time), as is apparent from our inspection of observations by Knopf et al. (2020).

We now make this derivation explicit in the paper by inclusion of a new Eq (1) about the time-dependency of the unfrozen fraction. However, we introduce two new features here: (1) an assumption that the relaxation time depends on time, as we infer from that inspection of the literature, and (2) the notion that drops which can *never* freeze during the isothermal experiment must be irrelevant to the time evolution of the unfrozen fraction, which should therefore exclude them in its definition.

We have added fresh text to clarify (new Eq (1) and lines 667-675).

Reviewer: Abstract: The abstract could be extended by a sentence that the authors have added to the conclusion section in the revised manuscript: “Any purely stochastic model of INP activity, assuming that the fractional freezing rate of all unfrozen drops is constant, would predict very high frozen fractions after a certain time, which would be inconsistent with our measurements. Instead, the statistical variability of efficiencies among INPs must be accounted for with any application of stochastic theory.” Or a similar statement.

Response: Done (lines 28-30).

Reviewer: Line 18: it should be mentioned that all samples were collected at the same station. The name of the sampling station could also be given.

Response: Done (line 18).

Reviewer: Line 75: “since INPs influence the ice concentration observed” instead of “since INPs determine the ice concentration observed” would be more precise since updraft velocity is also a major determinant of ice crystal number density in clouds.

Response: Done (line 77).

Reviewer: Line 76: do you mean here secondary ice production? If yes, it should be stated explicitly.

Response: This includes both SIP and homogeneous freezing.

Reviewer: Line 77: It is more than “beneficial” to simulate the first ice in mixed-phase clouds accurately. Consider to replace “beneficial” by “crucial” or something similar.

Response: Yes and no.

Naturally, it is necessary to predict correctly whether there is *any* onset of first ice. In some clouds (E.g. Sassen et al. 2003), subzero temperatures are too warm for any heterogeneous ice nucleation.

However, deep precipitating clouds will have an ice concentration that is determined by the inter-play between ice multiplication (defined as the positive feedbacks of microphysical processes involving SIP) and thermodynamic limits on the ice concentration (e.g. onset of subsaturation). So, an order of magnitude error in the concentration of the first ice may be irrelevant to overall accuracy of the eventual ice concentration for such clouds.

Some clouds are too thin for ice precipitation and hence for SIP too. For these, one needs to simulate the first ice accurately as it is the only ice.

We leave the text unchanged. “Beneficial” is nuanced and a fair compromise (line 79).

Reviewer: Lines 104–105: do you mean here that also the freezing rate should decline exponentially? Yet, for stochastic freezing, the freezing rate, i.e. the fraction of unfrozen droplets that freeze per time, remains constant. Just the absolute number of droplets that freeze per time decline together with the number of unfrozen droplets. In your terminology, the freezing rate seems to be termed a fractional rate. Then. it is absolutely unclear what you mean by “freezing rate”. As your terminology differs from the common terminology, there are many sources for confusion. If you want to stick to your terminology, it might be best to just remove “freezing rate” from the sentence.

Response: We agree.

There was confusion created by us using our own terminology that deviated from what appears to the common terminology.

For consistency with the common terminology, we now use “freezing rate” to refer to the fractional rate of change of the number of unfrozen drops throughout the paper.

Reviewer: Line 106: It should be added "for immersion freezing" after “This is seldom observed”, to make clear that this sentence does not refer to homogeneous ice nucleation, which is indisputable stochastic.

Response: Agreed. This is replaced as required (line 108).

Reviewer: Lines 259–260: SiO2, CaO, and Al2O3 are not minerals present in mineral dusts but the oxides that form after ignition of the samples, which is performed to determine the elemental composition. Mineral composition of dusts can e.g. be found in Murray et al. (2012), Kaufmann et al. (2016), and Boose et al. (2016).

Response: Agreed and text is changed as required with inclusion of these new references (line 266).

Reviewer: Line 290 and Table 1: The PM10 and ACSM derived aerosol concentrations are not always consistent. E.g. PM10 of the combustion dominated sample is 20.85 ug/m3, but the sum of Org, NH4, Cl, NO3, and SO4 adds up to 24.5 ug/m3. The reason for such discrepancies should be discussed.

Response: We have added a little more technical information about the PM measurements regarding size range and inlet heating in subsection 2.2.2.1 (lines 229-231).

At the end of subsection 2.2.2.2 (lines 337-342) we have added the following paragraph:

“In general, there was a clear tendency of the optically measured PM_1 to be lower than what was obtained from the summation of individual chemical components detected in the PM_1 fraction. We mainly ascribe the offset between the different measurement approaches to (i) the size range up to 0.18 µm not being detected optically, and (ii) the heated inlet before the optical measurements, which may lead to evaporation of (semi-) volatile aerosol particle components. The latter effect is likely to be more pronounced when nitrate species and (semi-) volatile organic species contribute significantly to the PM_1 (e.g. Huffman et al., 2009).”

Reviewer: Line 326: “aerosol components” might be a more appropriate wording than “aerosol properties”.

Response: Done (line 333).

Reviewer: Lines 473 – 477: this discussion is quite confuse and should be formulated clearer.

Response: We agree the paragraph was confusingly over-written.

It has now been re-written (lines 485-500).

Reviewer: Table 1: Bio Trak OPC showed elevated number concentration during the dust event from February 20 to 26. As mineral dusts can also be fluorescing, the Bio Trak signal should not be directly identified with PBAP in the presence of mineral dust as it is done in Table 1.

Response: Agreed. The heading in the table has been changed accordingly to "Fluorescing particles" instead of "PBAP". The caption has been modified (lines 1591-1594).

Reviewer: Lines 492–494: what is meant by "relatively small"? Please specify. What is meant by "a likely candidate"? The marine biogenic components? If yes, it should be "candidates".

Response: We have included these changes (lines 518-519).

Reviewer: Line 563 and Figure 8: it might be more meaningful to indicate the min–max value range rather than one standard deviation.

Response: We have updated Figure 8 to display the min-max value range for each drop instead of the standard deviation. We have also updated the text (lines 583-589).

Reviewer: Line 579: again, it might be meaningful to also state the min–max difference.

Response: We have added the requested information in more detail in Table 2.

Reviewer: Line 594–596: the definition of fice(t*) seems imprecise. More precisely, it should be "the fraction of droplets freezing starting from the isothermal phase (i.e. f_fice(t*=0) = 0).

Response: There is a misunderstanding here. f_ice is the total number of drops frozen since the start of cooling at 0 degC (when t* < 0). f_ice is zero at 0 degC and is non-zero when the isothermal temperature is first reached (t* = 0).

We have modified the text to clarify (lines 620-622).

Reviewer: Line 605: The fractional rate of freezing of unfrozen drops is the freezing rate! Please change accordingly.

Response: Text is added to clarify (line 632-636). We now call it the 'real freezing rate'.

Reviewer: Line 606: should this be fice(t*)/dt* as on line 603?

Response: The derivative is the same quantity, whether written as d fice(t*)/dt* or as dfice/dt*.

We now include the functionality '(t*)' in the written expression, to avoid confusion (line 634).

Reviewer: Lines 606–608 and Figure 11b: the presentation of the data in terms of the freezing fraction per fraction of unfrozen drops makes more sense than the presentation per fraction of frozen droplets, as the former represents the freezing rate, which should remain constant for stochastic freezing. It should be discussed why this quantity increases again for times since the start of the isothermal phase larger than 10^3 s for most samples. Also check for correctness as this behavior seems inconsistent with Fig. 9. Maybe discuss the uncertainty of the data points. Was this evaluation done for the averaged (yellow) data points shown in Fig. 9? If yes, you should consider smoothing them before analysis. As it seems, their scatter is increased compared with the individual datasets through averaging the datasets.

Response: We agree.

We now see that in the previous version of the paper, the lack of decrease of the fractional freezing rates after half an hour was due to a "noise floor" at long times due to too few raw data-points determining the plotted yellow points in Fig. 9 (averages among experiments in a sample) and increased random variability.

We have followed the reviewer's suggestion and now have included extra smoothing to eliminate any decrease in frozen fraction with time, before computing the derivatives for Figure 11.

The new Figure 11 is included, showing a steady decrease with time in the real freezing rate throughout the isothermal period, without such a noise-floor (lines 636-637).

Reviewer: Lines 614–615: what chemical kinetics are meant here? This should be explained.

Response: Done (lines 644-646).

Reviewer: Line 616: what is the "natural time scale of freezing"? This should also be explained.

Response: Done (line 648).

We now write that "*The reciprocal of the real freezing rate is the natural time scale of the freezing at any instant*".

Reviewer: Line 626–627: this sentence is confusing. Do you mean: "such less efficient drops" instead of "such less efficient INPs"?

Response: No, we meant "such INPs that are less efficient" at nucleating ice.

The sentence is now modified to clarify (line 657).

Reviewer: Lines 654–657: Making the fraction of droplets freezing per time interval a function of the unfrozen droplet fraction would indeed be a physically more meaningful formulation. If it were formulated like this, 1/tau(t*) could indeed be viewed as the probability of unfozen drops freezing during the isothermal phase. Consider to revise the manuscript in this respect.

Response: 1/tau always was that probability of unfrozen drops freezing during the isothermal phase. We show this now with an extra Equation (a new Eq (1)) which is the basis for the same isothermal formulation as before (now Eq (2), previously Eq (1)).

Text is added to explain the new Eq (1) (lines 667-675).

Reviewer: Line 709: what is meant by "somehow"? Please specify.

Response: We agree, the text was unclear and we now include some explanation of what we mean (line 747).

This assumed correspondence is discussed later, in Section 5. So we now include a cross-reference just there (line 747).

Reviewer: Line 710: the way the equation is formulated, the temperature shift in Eq. (4) applies to all INPs. Shouldn't it then be "applied to all INPs"? The sentence should be changed accordingly or it should be commented why only "most INPs".

Response: Yes, the term "all INPs" now replaces "most INPs" as required, and we add an extra sentence to clarify (lines 747-750).

The fixed shift downward is the approximation for the model; the shift upwards is what is real and is for the freezing temperature, which follows a statistical distribution instead of being fixed.

Reviewer: Line 831–832: this sentence is not complete.

Response: Now altered to clarify (line 873).

Reviewer: Line 964: Does the statement in the bracket refer to the treatment of temperature dependence suggested in this study? Please clarify.

Response: Yes, it is the modelled temperature shift for the INP scheme that is being referred to. It is now clarified (line 1007).

Reviewer: Line 974–976: this last sentence should be formulated in view of the discussion above, because it sounds as if the temperature dependence could indeed account for the observations of Westbrook and Illingworth.

Response: The sentence is now re-phrased to avoid implying that we expect a role of time-dependence (line 1019).

However, in light of ongoing simulations of cloud cases we now begin to wonder if the Westbrook and Illingworth hypothesis about this role just might be partly correct. "The jury is still out" on this question, as is now discussed in the text (lines 978-985).

A cloud simulation is needed to resolve this matter.

Reviewer: Table 2: it should be stated whether low or high numbers stand for uniqueness.

Response: We presume the reviewer meant “Table A2”. The caption is now modified to clarify.

Reviewer: Table B1: The parameter “Q” should be explained better in the list of symbols: “passive tracer of what?” What is meant by “freezing level”? Why does Q have units of kg[air]-1? On line 836, a value of Q is given without units.

Response: The problem with units is now corrected by introducing a new variable for the value of Q prescribed as unity outside the cold-cloud region. The text and equations are slightly changed accordingly.

Reviewer: Figure 9b: Axis labels in panels (b) need to be enlarged to the size shown in panels (a).

Response: We have updated the labels as requested.

Reviewer: Table 3: chi for the mineral dust influenced sample should be 1.48 instead of 1.5.

Response: No, the chi value is from the exact measured values, which are not displayed. Only two decimal places are displayed in the various columns.

Technical comments:

Reviewer: Line 59: The meaning of PK97 should be given here, at the first mentioning of Pruppacher and Klett, and not on line 91.

Response: Corrected.

Reviewer: Line 585: “time increases” instead of “times increase”.

Response: The sentence is now changed (line 610).

Reviewer: Table 4: the upper value of the continental pristine sample is not correctly displayed.

Response: Does the reviewer mean the extra decimal for the upper bound for the "Continental polluted" sample? ~~445.5~~ -> 446 ? Corrected.

Reviewer: Table 5: "°C" should be removed from "-16.3°C". The line numbers appear within the table.

Response: Done.

Reviewer: References:

Boose, Y., Welti, A., Atkinson, J., Ramelli, F., Danielczok, A., Bingemer, H. G., Plötze, M., Sierau, B., Kanji, Z. A., and Lohmann, U.: Heterogeneous ice nucleation on dust particles sourced from 9 deserts worldwide – Part 1: Immersion freezing, Atmos. Chem. Phys., 16, 15075–15095, https://doi.org/10.5194/acp-16-15075-2016, 2016.

Kaufmann, L., Marcolli, C., Hofer, J., Pinti, V., Hoyle, C. R., and Peter, T.: Ice nucleation efficiency of natural dust samples in the immersion mode, Atmos. Chem. Phys., 16, 11177–11206, https://doi.org/10.5194/acp-16-11177-2016, 2016.

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

Replies to Reviewer 2

Author response

We are grateful to the reviewer for their effort in scrutinizing the manuscript.

It is a fair criticism to make of the paper about the lack of certainty in attribution of INP types to some of the observed freezing behavior of samples. However, more error could be introduced into a model from treating all INP types with the same time-dependence treatment (same expression for the temperature shift) than for making use of the likely attribution noted below and in the paper.

Point-by-point Responses

Reviewer: General Comments

The authors have done a commendable job of addressing my points and those of other reviewers. I still very much appreciate this study overall, and wish to see it formally in the literature. I also appreciate the effort made to more fully characterize aerosol compositions involved in the classification of cases, to clean up the equations and make more precise explanation, and to add methodological details.

Response: We appreciate the encouraging and constructive style of the review.

Reviewer: While the aerosol chemistry does clearly indicate differences between the cases, and the statistical differences of some of the INP populations is supported, my opinion is that it is no clearer that such categorizations tag reliably specific INPs or makes them any more meaningful for categorization in models. This is acknowledged in the paper. Nevertheless, I think it is entirely possible that in follow-ups to this study, **the authors or other authors will use these categories or consider the polluted or high black carbon or high dust cases as representative of more specific INPs categories in numerical models. I think that would be in error, as more effort is needed** in future research to get to the point of a true "closure" study.

Response: We disagree that such a future initiative would constitute an "error".

We have provided much experimental data characterizing the various airmass types and their freezing behaviours in our experiments in the present paper.

It is curious that the review provides no evidence to falsify the likely attributions we have made about dominant INPs in the various samples (e.g. the mineral dust influenced sample we assumed to be probably reflecting the freezing by mineral dust INPs).

We now have included extra text with more analysis of our own chemical characterization to indicate likely INP species dominating the freezing of the samples (lines 485-504).

Creative modellers do not have the luxury of waiting years for perfect lab data to become available, as noted already.

Reviewer: This regardless of the apparently noble efforts modelers are making to deal with imperfect observational data, to paraphrase one of the more ludicrous responses I have seen (i.e., modelers not having the "luxury" to wait).

Response: **Our responses were fine. It is difficult to respond to such a tangential negative comment in a review when no coherent argument is provided.**

Regarding our philosophical comment about modellers not having the "luxury" of waiting years for perfect empirical data to become available, it was quite reasonable.

A key difference between the activities of state-of-the-art modeling and experimental observations is that any modeler must show that their model is realistic before using it for scientific questions. That involves comparing the model with observations of the real phenomenon being simulated, which consists of many processes and is to be represented somehow in all its complexity. The toughness of that challenge is why the modeler cannot wait for perfection of published experimental results when faced with any given incompletely characterized process during development of the model. The modeller must attempt some representation of it, since not to do so likely introduces more bias in the overall simulation of the wider phenomenon than use of albeit imperfect data.

This is a perpetual dilemma that modellers will always face, one could argue. Two of the co-authors are modellers, so we speak from experience. In fact, this dilemma is in a sense a reason for the current project happening in the first place, about how to treat time-dependence in a cloud model. When planning the project, we knew there would be such difficulties in the lab observations and that any data we would acquire would be incomplete.

It goes without saying that in the above comment, we are not referring to the simplest modeling that involves little validation or development of a model, or perhaps uses a model already created elsewhere.

By contrast, observationalists can focus on characterizing a single process in any study, or can simply record their observations of a complex phenomenon. They face no such necessity to address many processes simultaneously.

Of course, we are not saying that generally creative modeling is more challenging than observations overall. The nature of the difficulties differ between both types of scientific activity.

Reviewer: It is well known that biological/biogenic INPs play a significant role in the atmosphere at temperatures >-20C, and also that they are the most difficult category to quantify and pin down. Over a land location especially, these must be present at some level always, and they cannot simply be characterized by total biological particle concentrations from a given sensor. INPs are always more specific, even within some broad categories, and this is likely especially true for the most efficient ice nucleators that are active at modest to moderate supercooling.

Response: Agreed.

Even if one could measure the concentrations of individual types of bioaerosol (fungal, bacterial, pollen…) in a given ambient aerosol sample, as indeed we did a recent study (Patade et al. 2020), a grave

experimental obstacle is that in the freezing experiments it is uncertain which aerosol types from the immersed sample caused drops to freeze. It is not an insuperable obstacle but would take much effort to overcome.

Reviewer: In the end, I think that readers will focus most on the quantification and possible generalization of time dependence for ambient INPs (i.e., the focus of the title and what is in the abstract), and will focus less on the classifications of cases that represent some unknown mix of different INP types in all cases. I list a few specific points below for possible attention.

Response: Perhaps.

Reviewer: Specific Comments

1) In the new discussion circa line 942, is it necessary only to point out the utility of findings only for INP measurements that have short residence times (i.e., real-time measurements like a CFDC)? One imagines that it means that slow cooling or isothermal measurements, while helpful for validating results such as presented in this paper, are overall not necessary even for classical immersion freezing measurements.

Response: This comment seems rather subjective. The paper has not proven that time-dependence is unimportant for all clouds. Thin stratiform cloud without much SIP, as observed by Westbrook and Illingworth (2013), might be affected by time-dependence of ice nucleation somehow, because it lacks SIP. We are trying to ascertain this by simulations now for another project.

Reviewer: 2) The authors note that they found weaker time dependence across cases than found in the literature, and they reference Herbert et al. (2014) as supporting that different INP types should show little difference. That is not a result highlighted in Herbert et al., as far as I read that paper, and it is also the case that it was heavily focused on inorganic materials, especially minerals.

Response: This comment prompted us to re-read the Herbert et al. paper and we now agree there was a problem with the way we cited it. Herbert *et al.* never wrote that the degree of time-dependence is independent of INP composition.

The citation of Herbert *et al.* has now been corrected (lines 128-131).

Reviewer: Also, Wright et al. (2013; doi:10.1002/jgrd.50365), Fig. 3, shows that some types of INPs vary in time dependent character when isolated. But this all leaves me to **wonder, again, if there is any reason to think that the different cases are representative for future application to specific INP scenarios, versus a more generalized time dependence to use for any INP category** (at least to the extent that data at -15C characterizes things across temperature of relevance to mixed-phase clouds)?

Response: Yes.

We agree that the degree of time-dependence varies with INP composition.

We see the various samples display differing degrees of time-dependence, which would reflect different INP types. Also, the higher the initial frozen fraction, the lower the time-dependence and vice versa.

So we think that the various cases are likely to be representative of specific INP types, the identity of which is uncertain for our samples. Our mineral dust influenced sample displays a stronger time-dependence than the other samples, consistent with Herbert et al. also observing a stronger time-dependence (montmorillinite). There are reasons to suppose that the IN activity of the rural continental sample is likely influenced by PBAPs, since other components are low, and since the temperature gradient of atmospheric INPs is weak towards the warmest temperatures in Fig. 6, where PBAP-IN are uniquely active, (lines 500-504 and 859-863).

However, we also argue that the differences in the degree of time-dependence observed among samples are rather limited. So, lack of certainty in inferring the dominant INP type for each is not a prohibitive problem for modeling overall effects from time-dependence, as we now say in the text (lines 863-865).

Reviewer: I know that categorization to fit model categories was an imagined goal of this paper, but it remains the one that has the least clear support. I would even say that the careful effort put in to attempting to categorize aerosols and air mass characteristics as related to INPs stands as testament to how extremely difficult it is to characterize INP scenarios, since INPs are but a limited fraction of the entire aerosol population. The INP data are certainly representative for the region, but parsing them out to different sources is not possible from the data collected. The story on time dependence is the true feature result here.

Response: Well, the mineral dust influenced sample originates from the Sahara and the degree of time-dependence differs from say the continental pristine sample. Mineral dust is the most prolific INP type active at the temperatures we studied (near -15 degC) in the background troposphere. So, it is likely that the freezing behavior for that sample reflects that of mineral dust IN.

Similarly, there are reasons to suppose that the rural continental sample may have active INPs influenced by PBAPs, partly since it was taken in a warmer season and due to the measured composition of low amounts of other components (lines 500 and 863). Also, its active atmospheric IN seems enhanced at the warmest subzero temperatures (see the gradient for the rural continental sample in Fig. 6). PBAP-IN are often identified by their activity at uniquely warm temperatures.

Reviewer: 3) Regarding new Fig. 5 and discussion around it (much appreciated), background is important whether in the temperature regime where it is “rare” or where the frozen fraction gets very high. The question I have, and which needs a clear statement, is if any corrections are actually applied, and if the rare occurrences at higher temperatures are simply ignored. I am not judging, just saying that

it needs to be said. Some would average all background cases and do corrections, but one at least needs to say what was decided.

Response: On average, we observe a frozen fraction of 0.01 for a temperature of -20°C for the ultra-pure water experiments presented in Fig. 5. As can be observed from Fig. 7, the frozen fraction was typically between 0.8 and 1 at a temperature of -20°C for the ambient samples. Hence, if the substrate potentially could induce a very minor frozen fraction around that temperature – it is a lot more likely that INPs in the samples already induced freezing at a higher temperature in those few potential cases. Thus, we find no reason to correct for that background in the results presented in Fig. 6 – and any such minor correction would not influence the presented results.

We would not expect any low-quality slides to bias the presented results significantly since we observed very high reproducibility between different droplet populations for the same sample (Fig. 7).

The following statement has been included early in section 3.1.1: "The presented INP spectra have not been corrected for the background, as the background was negligible relative to INP concentrations in the ambient samples." (lines 466).

Reviewer: 4) The data availability statement is not up to current standards and expectations, in my opinion

Response: The statement has been improved.