Response to the Comments of Reviewer 2

We would like to thank Reviewer 2 for his comments and thoughtful suggestions. We reply to the individual comments below.

General comment *First of all, this is a very impressive set of measurements. The field of atmospheric ice nucleation is lacking long term data sets with which to compare models and test our understanding. Hence, a dataset comprised of 500 individual measurements over the course of 5 years is extremely valuable. Hence, I support its publication. However, I think there are some aspects of the paper which need to be improved prior to publication. I go through these in detail below:*

Specific comments

1. *P1. Ln 11. Why does an exponential change in INP concentration suggest that several groups of particles with different ice nucleating properties are contributing to INP populations? The exponential dependency on T could also just be explained by a distribution of sites across a surface, rather than particles of different composition or size.*

   The crucial point in our view is that the INP concentration is observed to exponentially increase by seven order of magnitude from -5 °C to -38 °C i.e. the steepness with which the concentration increases on a log-scale. This increase is shallower than the increase in ice nucleation activity of any pure substance, bringing us to the conclusion that several substances contribute to the ambient temperature spectra. To clarify, we rephrase in the manuscript: "Concentration of ice nuclei is found to increase exponentially by seven order of magnitude from -5 °C to -38 °C. Sample to sample variation in the steepness of the increase indicates that particles of different origin, with different ice nucleation properties (size, composition) are contributing to the ice nuclei concentration at different temperatures."
   Our interpretation of the temperature spectra is a central point of criticism of Reviewer 2, also in comment 7, 8 and 12. We attempted to illustrate our point of view in Appendix A in the manuscript. Below we try to clarify and support our conclusions by following the Reviewers suggestion to use active surface site distributions to explain the observed temperature spectrum. We show how the atmospheric INP spectrum compares to an INP spectrum that is constructed using concentrations of ice nucleation active surface sites (referred to as INAS or n_s) for desert dust and for Microcline.
   The temperature spectra (INP concentration as function of temperature) generated by a substance is the substance's ice nucleation activity (e.g. frozen fraction as function of temperature) multiplied by the number of particles of this substance. We assume one particle per droplet.
   Frozen fractions can be derived from n_s parametrizations by $FF = 1 - exp(-n_s \cdot A)$ with A being the particle surface area per droplet. The frozen fraction envelop in Fig. 1(a) is obtained by using A of spherical particles with diameter between 100nm and 1μm.
   Fig. 1(a) shows the calculated frozen fraction for n_s parametrizations, found for the K-feldspar, Microcline (Atkinson et al., 2013) and different desert dusts (Ullrich et al., 2017). Microcline is considered the most ice nucleation active dust identified so far (Atkinson et al., 2013), we come back to that in comment 7. Desert dust is not a pure substance but a mixture of different minerals and whatever sticks to the mineral dust particles (e.g. biological residues, Conen et al., 2011).
   Looking at Fig. 1(a) it can be seen that the ice nucleation activity of both, desert dust and Microcline is higher than that of a hypothetical substance that would correspond to Fletcher's curve. As pointed out by the Reviewer, using n_s results in an exponential dependency on temperature. However the slope of Microcline is steeper than the Fletcher curve. Based on laboratory measurements on other, seemingly pure substances (e.g Fig.1a, Atkinson et al., 2013) we argue that all pure mineral dusts exhibit a comparable or steeper slope than Microcline, i.e. activity increases more rapidly with decreasing temperature than that of a hypothetical substance that would correspond to the Fletcher curve. As a consequence, if only one mineral dust would contribute to the concentration of INP in the entire temperature spectra, the spectrum must have the same steepness as the frozen fraction of that mineral. Only Microcline, as an example, would produce the temperature spectrum shown in Fig. 1(a) with the right hand y-axis (calculated by $FF_{Microcline} \cdot N_{IN@233K}$). While INP concentrations between 263-268K would correspond to observational data, at 253K where typical INP concentration is ~$1\ell^{-1}$,

Microcline would produce ~1000ℓ^{-1}. This is not what is observed. The lower temperature dependence of the frozen fraction calculated using n_s of desert dust must result from a superposition of several steeper curves of all substances composing desert dust in their specific fraction.
Fig. 1(b) shows the temperature spectrum from multiplying desert dust's ice nucleation activity with a globally averaged dust concentration given in Atkinson et al. (2013). This represents observations well, in steepness and range. As mentioned above, we argue that the match in steepness stems from the contribution of different substances contained in desert dust.

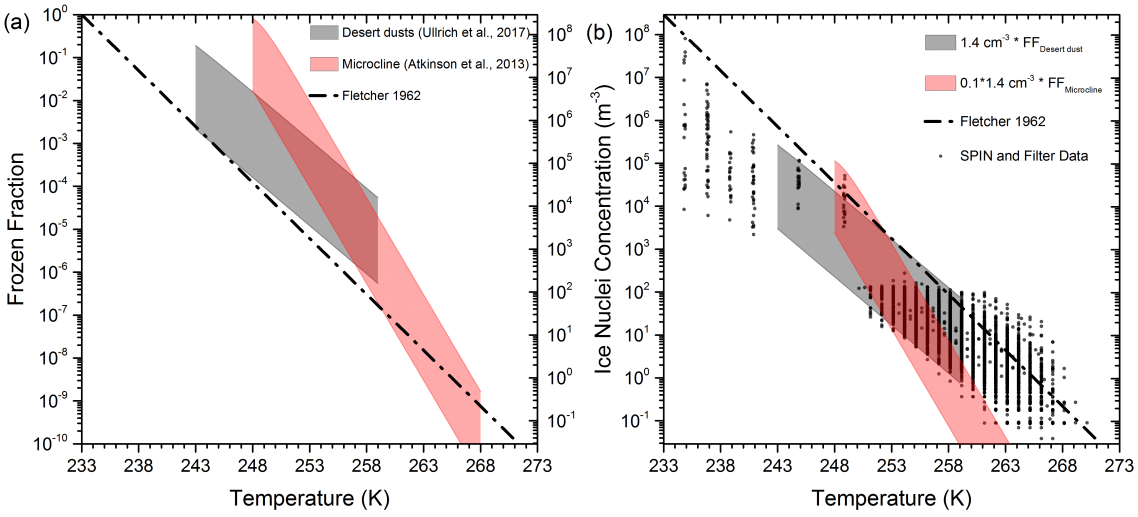

Figure 1. (a) Frozen fraction (left side axis) of of Desert dust, Microcline and ambient INP as function of temperature. Frozen fractions are calculated based n_s from Atkinson et al. (2013); Ullrich et al. (2017) for particle diameter between 100nm and 1μm, respectively by normalizing Fletcher's parametrization by the maximum predicted INP concentration (at 233K). Multiplying frozen fractions by the normalization factor leads to the corresponding ice nuclei concentrations (right hand axis).
(b) Temperature spectra including Fletcher's parametrization and n_s predicted concentration, scaled by globally averaged dust concentration for desert dust and 10% of globally averaged dust concentration for Microcline.

2. *Consider using the term 'ice nucleating particles; INPs' rather than ice nuclei (IN). Vali et al. [2015] in their recent definitions paper came up with some compelling reasons why this is a better and less confusing term.*

   As we are investigating filter samples and ice nuclei may be of various nature (particles, macro-molecules, ...) we rather prefer to stick to the classic abbreviation IN. We now state in the introduction that IN stands for "ice nucleating substance".

3. *P1 ln 24. Provide a citation for 100 L-1 INP leading to diamond dust. My understanding was that diamond dust was a relatively low concentration of ice crystals of relatively large size, i.e. in contradiction to the statement made here. In addition, I understand diamond dust tends to form in clear air, without the presence of a liquid cloud.*

   The sentence is inspired by the following report by Vincent Schaefer: "One late winter day in 1944, when I was climbing Mt. Washington by way of Tuckermans Ravine with Dr. Irving Langmuir, we approached the base of an orographic cap cloud which covered the cone of the mountain. He pointed out that the concentration of ice nuclei in the cloud was probably less than one per 1000 cubic meters, i.e. 1 x 10^{-6}/liter! He made his estimate using the horizontal and vertical distances of about 10 meters existing between the snow pellets which were falling from the cloud. We were also aware that at other times, what we call diamond dust, could be observed under similar temperature conditions with

concentrations as high as 100 per liter." (Schaefer, 1967).
The Reviewer is correct that the connection to ice formation in liquid clouds is not obvious without context. As no firm evidence could be found elsewhere, the sentence is removed from the manuscript.

4. *P2, ln 1. When discussing data like that of Ansmann et al. and making statements such as 'above -10 C ice containing clouds are rare', make sure it is stated what sort of clouds are being referred to. For example, in convective clouds ice formation above -10oC is common. Ansmann et al. deal with shallow clouds.*

   Ice formation via immersion freezing should occur at the same temperatures, independent on the dynamical conditions under which clouds form. The difference must be due to secondary ice formation, which is more effective in cumuli than in stratiform clouds.
   The cloud type in the studies cited is added to the manuscript.

5. *P2. Ln 3. This paragraph is very confused. Parts of it seem to be referring to ice formation in shallow cloud types (e.g. stratus), whereas it then morphs into a discussion about secondary production which is more relevant for deep clouds.*

   We have restructured and extended this part of the introduction to make it more organized. The cloud regime in which the measurements we refer to have taken place is indicated.
   It now reads: "Ambient measurements of IN-concentrations from various studies were compiled by Fletcher (1962) to derive a spectrum of the average IN-concentration as a function of temperature (curve shown in Fig. 1). Fletcher's curve has been found to match ice crystal concentrations measured in stratiform clouds and cold-based convective clouds but underpredict the concentration in deep convective clouds (e.g. Mossop, 1985; Cooper, 1986). Matching concentrations of ice crystals and IN indicate a direct influence of immersion freezing IN on cloud properties.
   At what concentration ice crystals exert a substantial influence on the properties of the cloud in which they form, has been addressed by Rangno and Hobbs (1988). From aircraft and mountaintop observations, Rangno and Hobbs (1988) identified the significant ice crystal concentrations able to produce precipitation to be on the order of $1\,\mathrm{L}^{-1}$ or more, cf. Fig. 1. In their data-set, such concentrations have been measured in cumulus clouds with top temperatures between -5 °C and -10 °C. At this temperature, this is a much larger concentrations than expected from the Fletcher curve and indicates secondary ice formation. Secondary ice formation mechanisms (most efficient at -5 °C) can increase the ice crystal concentration within a cloud by as much as four orders of magnitude above the number of IN present (e.g. Hallett and Mossop, 1974; Hobbs and Rangno, 1985; Mossop et al., 1970). Already few active IN ($0.01\,\mathrm{L}^{-1}$) can be enough to start the multiplication (Sullivan et al., 2017). In-situ evidence for secondary ice formation is reported in Hoffer and Braham (1962). They collected graupel from the top of cumulus clouds to melt and refreeze them under laboratory conditions. They found that every sample froze at substantially lower temperatures than the lowest temperature in the cloud from which they were collected, indicating that they did not contain IN active at the temperature the ice pellets froze in the cloud.
   Typical temperatures at which ice crystals are observed to form in numbers high enough to affect the properties of clouds have been measured by remote sensing. Satellite observations, averaging clouds globally (e.g. Carro-Calvo et al., 2016) and ground-based Lidar measurements, looking at shallow clouds (e.g. Ansmann et al., 2009), report that above -10 °C ice containing clouds are rarely detected and often cloud top temperatures below -20 °C are necessary for clouds to glaciate. Satellite data (Carro-Calvo et al., 2016; Rosenfeld et al., 2011) and aircraft observations (Rangno and Hobbs, 1991, 1994) agree on a land-sea contrast with the tendency of cloud glaciation at higher cloud top temperatures over sea. Carro-Calvo et al. (2016) offer the explanation that the presence of larger sized droplets in maritime clouds, which are required for effective secondary ice formation (Heymsfield and Willis, 2014) could play a role. Hallett and Mossop (1974) suggested that marine cumuli contain large ice crystal concentrations for dynamical reasons. They usually have higher cloud top temperatures, therefore the contact of ice and supercooled droplet occurs at temperatures favorable for splintering."

6. *P2. Ln 17-25. This discussion of marine INP is lacking reference to some more recent literature on the subject, e.g.: [Burrows et al., 2013; McCluskey et al., 2017; Vergara-Temprado et al., 2017; Wilson et al., 2015; Yun and Penner, 2013]. I appreciate the effort made to go back to much older studies, but the new work also needs to be discussed.*

We thank the Reviewer for pointing us to this recent studies. We included some of them at this point of the manuscript. "More recently, laboratory investigations (DeMott et al., 2016; McCluskey et al., 2017), modeling (Burrows et al., 2013; Vergara-Temprado et al., 2017) and ambient observations (Wilson et al., 2015) showed that under certain conditions, IN from marine sources can be abundant enough to significantly contribute to the total IN population, particularly in high latitudes."

7. *P2. Ln 25. The statement that 'From laboratory experiments it is established that dust particles tend to nucleate ice below -20C whereas biological particles can initiate immersion freezing at temperatures up to -5 C' is wrong. I can point to numerous studies showing dust can nucleate ice at much warmer temperatures; e.g. [Atkinson et al., 2013; Niemand et al., 2012; Ullrich et al., 2017]. Modeling suggests that dust is important in many locations at much warmer temperatures than -20 C [Vergara-Temprado et al., 2017].*

   We changed the sentence to:'From laboratory experiments it is established that dust particles tend to nucleate ice efficiently below -20 °C whereas some biological particles can efficiently initiate immersion freezing at temperatures up to -5 °C (Murray et al., 2012).'
   Modeling INP concentrations with only Feldspar suffers on over-predicting concentrations at low temperatures and under-predicting concentrations at warm temperatures, in agreement with what is demonstrated in Fig. 1, comment 1. The temperature of agreement between model output and observation shifts with the feldspar number concentration.

8. *P2-3. It is not possible to distinguish between dust and bio INP on the basis of an inflection at -16oC. It is false to claim that such an inflection would give you information about biological INP. In making this statement the authors are assuming they know what the ice nucleating spectrum of dust is and also that they know that biological INP nucleate around -16C. Neither can be assumed or are correct. Biological material has a huge diversity in its nucleating ability. There are exceptional ice nucleating materials from specific fungal and bacterial species and much less active materials associated with marine biology. Also, the work of DeMott et al. [2016] and Wilson et al. [2015] suggest that the slope of INP vs T for marine INP materials is quite shallow, in contrast to what is stated here.*

   There might be a misunderstanding. As shown in Fig. 1 comment 1, mineral dust contribute at temperatures lower than -16 °C, therefore changes in abundance of dust shifts the temperature spectrum up and down, below this temperature. Any inflection at warmer temperatures can be attributed to other INP species (which don't need, but could be biological). It is more difficult to differentiate between them on the basis of the temperature spectrum at lower temperatures.
   Shallow slope of any material must be due to its inhomogeneity. The largest inhomogeneity in marine INP is probably the diversity in composition i.e. various particle species in different concentrations and with differing activities. Laboratory results pointing in this direction can be found in McCluskey et al. (2017). They report that experiments when a phytoplankton bloom was provoked, caused a response at various temperatures during different stages of the bloom, suggesting a diverse marine INP population.
   The section was slightly changed to make it clearer. It now reads:"Two main sources for IN in maritime air have been proposed. Long-range transported continental aerosol (mainly dust) suggested by Bigg (1973) or marine organic ice nuclei of biogenic origin aerosolized with the sea spray (Schnell and Vali, 1975; Rosinski et al., 1987). Aerosol from both sources have been investigated (e.g. DeMott et al., 2003, 2016, and references therein). They found that marine-sourced IN are less efficient than IN from continental sources and can contribute IN in a broad temperature range. From laboratory experiments it is established that dust particles tend to nucleate ice efficiently below -20 °C whereas some biological substances can efficiently initiate immersion freezing at temperatures up to -5 °C (Murray et al., 2012). Joly et al. (2014) demonstrated that the particles initiating immersion freezing in cloud water samples collected at Puy de Dôme consist to an increasing fraction of biological IN (identified by sensibility to heat treatment) towards higher freezing temperatures. They estimate 77% biological IN at -12 °C increasing to 100% at -8 °C.
   The different temperature range in which dust or biological particles act efficiently as IN, can generate a specific signature in the concentration-temperature spectra if there is a change in abundance (cf. Sec. 4 and Appendix A). Some biological particles and in particular bacterial IN of one source tend to exhibit homogeneous ice nucleating properties. They initiate ice formation in a narrow temperature range seen as step like increase in concentration in a temperature spectra (e.g. Murray et al., 2012). In contrast, dust particles activate in a broader temperature range (e.g. due to inhomogeneities in

composition, surface structure and the influence of particle size on nucleation efficiency) seen as exponential increase in concentration towards lower temperatures (Bigg, 1961). If a strong marine source of biological origin exists it can be expected to be detectable as an inflection in IN-concentration at temperatures above -16 °C where the ice nucleating fraction of dust particles is small."

9. *P4. Ln 5. It would be helpful to see the control fraction frozen curves as well as the fraction frozen curves for the samples. These control experiments look better than those reported by Conen et al., why is this? What has been done differently?*

   We don't know what makes the difference. Fig. 2(a) shows the frozen fraction curves from the reported samples in comparison to three measurements of each the used water, fresh filter and field blanks of filters treated, stored and handled the same way as samples but removed from the Digitel before sampling. The contribution from sampled aerosol to the frozen fraction can be isolated from the background contribution of water and filter material. According to the 'At least one Rule' in probability calculus, freezing is caused by the aerosol, the background or both. Treating the frozen fraction as probability for a droplet to freeze we can calculate the probability for a droplet not to freeze:

$$(1 - FF_{BG}) \cdot (1 - FF_{Aerosol}) = 1 - FF_{Sample} \tag{1}$$

   Solving for $FF_{Aerosol}$ results in

$$FF_{Aerosol} = \frac{FF_{BG} - FF_{Sample}}{FF_{BG} - 1} \tag{2}$$

   where FF_{BG}, $FF_{Aerosol}$, FF_{Sample} denoting the frozen fraction only from the background, the aerosol and the measured frozen fraction of the sample. With FF_{BG} and FF_{Sample} known, Eq. 2 can be used to calculate the contribution which can only be attributed to the sampled aerosol. The result is shown in Fig. 2(b).

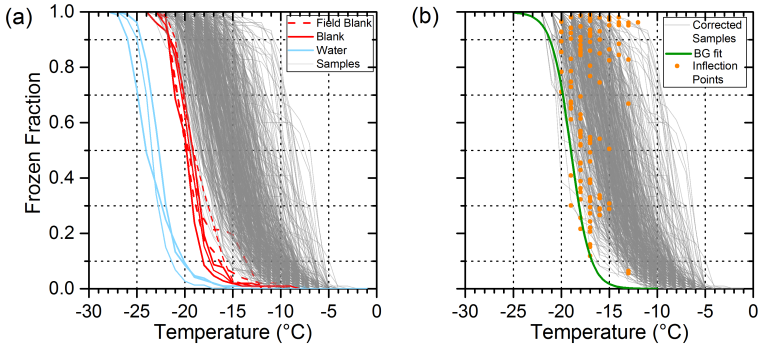

Figure 2. (a) Frozen fraction curves of filter samples (FF_{Sample}) in comparison to background measurements. (b) Minimal frozen fraction from sampled aerosol ($FF_{Aerosol}$). The FF_{BG} derived as a fit to blank measurements is shown in green. $FF_{Aerosol}$ curves are cut if their slope becomes negative and endpoints are marked in orange.

   For some measurements, isolating $FF_{Aerosol}$ in the way described above can lead to decreasing frozen fraction with decreasing temperatures. As this is unphysical, we cut such curves at their inflection point.
   The above analysis supports the temperature information given in the manuscript. We did not make changes to the text.

10. *Figure 1. Also show other INP parameterisations that are used in models in addition to Fletcher, e.g. Meyers et al, Cooper et al.*

    The two parametrizations from Cooper (1986); Meyers et al. (1992) have been added. In order not to overload the figure and distract from the actual measurements, no other additional parametrizations are shown. The temperature range of the

parametrization lines has been capped according to the validity range given in the original work. The parametrization lines are calculated according to Eqs. 3, 4, 5 and shown in the range of supercooling (ΔT) indicated:

$$\text{Fletcher 1962:} \quad N_{IN}[m^{-3}] = 10^{-2} \cdot exp(0.6 \cdot \Delta T), \qquad 10^\circ < \Delta T < 30^\circ \quad (3)$$

$$\text{Cooper 1986:} \quad N_{IN}[m^{-3}] = 10^3 \cdot 10^{(-2.35-0.135 \cdot -\Delta T)} = 10^{(0.65+0.135 \cdot \Delta T)}, \qquad 5^\circ < \Delta T < 25^\circ \quad (4)$$

$$\text{Meyers et al. 1992:} \quad N_{IN}[m^{-3}] = 10^3 \cdot exp(-0.639 + 0.1296 \cdot (100 \cdot (S_i - 1)))$$

$$= 10^3 \cdot exp(-0.639 + 0.1296 \cdot (100 \cdot (S_w \cdot \frac{p_{w,sat}(T)}{p_{i,sat}(T)} - 1))), \qquad 7^\circ < \Delta T < 20^\circ \quad (5)$$

11. *P10, ln 15-23. In this discussion of the conclusion that the authors see no evidence for marine INP, they need to cite other papers with similar conclusions. For example, Fig 5 of Vergara-Temprado et al. [2017] clearly shows that desert dust is much more important than marine organic INP in the Eastern Atlantic region. Similarly to the final statement referring to Burrows, Wilson et al. [2015] also conclude that marine INP might be important in the southern ocean. They do not make this conclusion on the basis that marine organics are particularly good at nucleating ice, they conclude this because the southern ocean atmosphere has very little desert dust in it and marine organics therefore define INP population.*

    We added Vergara-Temprado et al. (2017).

12. *P11, ln 15, Why are INP above -10 biogenic? This statement needs to be expanded upon or altered. As mentioned above, mineral dust can nucleate ice in this temperature regime.*

    In the course of improving the conclusion section the statement was deleted. However, we think there is evidence that it is true. See comment 1. We conclude this based on the observed INP-concentration, which is the product of activity and abundance of the substance. The activity of mineral dust at -10 °C is low and a high number of particles would be necessary to produce the INP-concentration observed in the atmosphere. At lower temperatures, the high abundance of dust particles would result in much more INP than observed (see the calculation example in comment 1 and Fig. 1b). This leads to two possible explanations. First, the explanation offered in our paper, that what makes the temperature spectra are the concentrations of different substances contributing in a temperature range where their activity is high . As the most active mineral dust known today is not active enough to explain the observed INP concentration at $T > -20\,^\circ\mathrm{C}$, another substance must be nucleating ice. Because Microcline is the most active mineral dust, no other mineral dust can be this more active substance. Natural substances with a higher activity are found in the class of biological aerosol. Alternative, there is a more active, very rare (low concentration) mineral dust that has not jet been identified or an entirely different source.

13. *P12. In this discussion of Ansmann et al., make it clear that this -20C number is for shallow clouds only, not deep convective clouds. In contrast the OSCIP from Rango and Hobbs is for cumulus clouds. Consequently I think the link between these ground level measurements and mid-level clouds is not as clear as the authors suggest.*

    Ansmann et al. (2009) measured alto cumulus clouds. This is now mentioned in the manuscript. The evidence is circumstantial, but the point is that an ice crystal concentration of $1\ell^{-1}$ is a benchmark concentration to clearly change mixed-phase cloud properties. In clouds where primary ice nucleation dominates ice crystal formation, this concentration is observed at -20 °C in agreement to our measurement of the INP-spectra at ground-level. We think this is an interesting agreement and kept it in the text. It could also be evidence that concentrations measured at ground are similar at cloud level.

14. *Conclusions: Some of these paragraphs are very short and there is a single sentence paragraph which seems to be floating and not connected to other statements. Hence, it reads more like a list of bullet points than a well-crafted conclusions section. This could be improved.*

We re wrote and condensed parts of the conclusion. It now reads:
"Typical IN-concentrations in the subtropic, marine environment are obtained from 500 particle filter samples collected over 4 years and from a 2-month field campaign with the SPIN instrument. Concentrations increase exponentially from $10^{-1}\,\mathrm{m}^{-3}$ to $10^{6}\,\mathrm{m}^{-3}$ between -5 °C to -38 °C. Over time, the concentration varies up to three orders of magnitude on a synoptic scale (filter samples) and up to four orders of magnitude on a higher resolution (SPIN measurements). The frequency with which certain concentrations are measured at a temperature T follows a log-normal distribution, characteristic for successive random dilution during long-range transport. The log-normal frequency distribution is found at all investigated temperatures. Parametrizations in numerical models should reproduce this feature.
For the long-term series of filter measurements, we find that random dilution during transport could account for larger fluctuations in IN-concentration than seasonal changes, changes in air mass origin and changes in the bulk aerosol composition. At Cape Verde, the later do not reveal a clear correlation to the measured IN-concentration. The only observable tendency is that continental air mass contain higher IN-concentrations than maritime air mass, in agreement to previous measurements (e.g. Bertrand et al., 1973) and model predictions (e.g. Burrows et al., 2013). The absence of an annual trend despite a dust and a biological active season is surprising and highlights the lack of knowledge about the nature of ice nucleating particles.
The diversity of the ice nuclei population manifests in the shape of the cumulative temperature spectra. The contribution of each source to the temperature spectra depends on the source strength, dilution during transport and the temperature range to which the source contributes efficient IN. Multiple sources, together with random dilution, could produce the appearance of an ubiquitous, almost constant background concentration of IN.
Assuming $10^{3}\,\mathrm{m}^{-3}$ to be a threshold IN-concentration to have a significant, primary effect on properties of supercooled clouds, temperatures below the range covered by the drop freezing experiment used in this study, need to be reached to have an effect. If the temperature dependent IN-concentration exponentially increases with the slope of the Fletcher (1962) approximation, a primary effect of ice nucleation by immersion freezing on cloud glaciation and precipitation formation can be expected at $-20\,^{\circ}\mathrm{C} \pm 5\,^{\circ}\mathrm{C}$, matching the -20 °C alto cumulus cloud glaciation temperature over Cape Verde reported by Ansmann et al. (2009). The SPIN data measured at temperatures below -24 °C support the temperature trend. Given that measurements of IN-concentrations at ground are representative for higher altitudes, the observation of cloud glaciation at $-20\,^{\circ}\mathrm{C}$ where IN-concentrations reach OSCIP support the importance of immersion freezing on cloud properties in a subtropical, marine environment."

Technical corrections
None.

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