Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., author comment AC2
https://doi.org/10.5194/acp-2021-700-AC2, 2022
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Reply on RC2

Christian Tatzelt et al.

Author comment on "Circum-Antarctic abundance and properties of CCN and INPs" by Christian Tatzelt et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-700-AC2, 2022

Answers to Reviewer 2

Anonymous Referee #2, 17 Dec 2021

Summary

RC2.1: Tatzelt et al. present results from detailed measurements of CCN and INP over the Southern High Latitudes during the Antarctic Circumnavigation Expedition (ACE). These measurements are extremely valuable to the field of polar aerosol-cloud interactions. The authors include a discussion of how these measurements compare with previous measurements of CCN and INPs and also perform correlation analyses with co-located aerosol metrics in aims of determining the source origin of measured CCN and INP abundances and variability. Overall, there is massive value in these measurements and the manuscript is well written with few typos. However, there are many studies in the literature that have not be considered in the interpretation of the INP data that I think will add significant scientific value to the discussion. Additionally, I found several statements that were not adequately supported with data. As such, I recommend publication in ACP after the authors address the major concerns identified below.

AC2.1: We thank the reviewer for the positive and constructive review. Their comments made our study much more targeted, structured, and understandable. We have therefore rewritten the text following the major and general comments below.

Major Comments:

RC2.2: The reported INP number concentrations were compared to the Bigg 1973 measurements. However, more recent measurements reported by McCluskey et al., (2018) indicated up to 100 times lower INPs compared to the Bigg 1973 survey. Many, perhaps the majority, of INP number concentrations in this study are significantly higher than those reported by McCluskey et al. (2018) and more recently McFarquhar et al. (2021), which is interesting. The more recent data is only briefly mentioned and discussed in comparison to the Bigg survey. These ACE measurements should be discussed in the context of both Bigg (1973) and McCluskey et al. (2018) datasets for completeness.

AC2.2: Please see AC1.4. We have now included a detailed comparison to McCluskey et al. (2018).

RC2.3: The authors indicate that an assessment of air mass origin is not possible for the ACE observations due to a lack of a tracer measurement (lines 403-404). Given the difference in these measurements and those reported in McCluskey et al. (2018) and more recently McFarquhar et al. (2021), it would be extremely valuable to determine if the air masses originated from open ocean waters, the Antarctic coast, or one of the surrounding land regions. While dust concentrations are extremely low over the Southern Ocean, it is widely recognized that the ice nucleation ability of dust is significantly greater than sea spray aerosol (DeMott et al., 2016) and so even small amounts can significantly influence the measured INP. Dust sources have been identified for Antarctica (e.g., Neff et al., 2015). Have the authors considered using something like HYSPLIT back trajectories to determine air mass origin?

AC2.3: Backward-trajectories for ACE are available in Thurnherr et al. (2020, https://zenodo.org/record/4031705) and we now include an analysis of the air-mass origin in comparison to N_{INP} from LV filter samples. We added a detailed description of the methods to the SI. Results are shown in the new Fig. 7. Overall, this analysis helped to underline our findings regarding the origin of the two distinct modes in the PDF for $N_{INP,-12}$, $N_{INP,-16}$, and $N_{INP,-20}$ in Fig. 8b—d. However, there is no clear signal in INP abundance from air-masses passing over Antarctica (Fig. S7b).

L465 tracked changes: *"This assumption is supported by the results of the air-mass origin analysis (subsection 2.4) using the LAGRANTO backward trajectories for ACE provided in Thurnherr et al. (2020). An overview of the results is given in Fig. S6 showing time series of surface contributions to each LV filter. The time series for the surface type contributions to the PBL signal (Fig. S6c) and the contribution of geographical regions (Fig. S6d) show that periods of elevated INP concentration (Fig. 5) coincide with periods when air-masses that passed over African, Australian, South American land masses or coastal regions were sampled. Contrary to these regions, air-masses passing over Antarctica did not show higher* N_{INP} *than oceanic air-masses (Fig. S7)."*

L517 tracked changes: *"The air-mass origin for the whole cruise and individual Legs are presented in Fig. 7. The average contributions for the whole cruise are dominated by air-masses from the open ocean, with contributions of at least 80 % and up to 97 % (Leg 1). The terrestrial air-masses (land and coast; excluding Antarctica) contribute only between 2 % (Leg 1) and 12 % (Leg 3). Similar Leg-wise average contributions could, hypothetically, be a result of dominant contributions of "open ocean" conditions during all Legs combined with limited INP variability over the entire SO for "open ocean" conditions."*

L551 tracked changes: *"This mode has a greater terrestrial and coastal influence (combined □35 %) than the low concentration mode, based in the air-mass origin analysis (Fig. 7). In consequence, the high concentration mode is labelled "terrestrial/coastal" in Fig. 8b–d (yellow area)."*

L782 tracked changes: *"The PDF for* N_{INP} *(T) shows two concentration modes at −20, −16, and −12°C (Fig. 8). The analysis of backward-trajectories indicates that low concentrations are associated with air-masses from the open ocean and from Antarctica, while the air-masses transporting higher* N_{INP} *passed over (non-Antarctic) land."*

RC2.4: The authors reference the DeMott et al. (2010) INP parameterization briefly. However, studies (e.g., Vergara-Temprado et al., 2017) have found limitations of DeMott et al. (2010) for marine regions due to the fact that the data used to develop the DeMott et al. (2010) parameterization did not include marine data. Why have the authors not included a comparison against existing marine INP parameterizations based on total aerosol surface area (McCluskey et al., 2018) aerosol volume (Mitts et al., 2021), which could be tested against the measured aerosol distributions from the merged size distributions?

AC2.4: References to these parameterisations are now included. Number site density and volume site density have been calculated and added as Fig. S8b and Fig. S8c. The text has been changed, accordingly (see below).

L595 tracked changes: *"In order to test the hypothesis that typical marine INP (e.g., SSA) were encountered for the majority of the cruise, concentration values were normalised. This was achieved by dividing* N_{INP} *by the particle surface area concentration or the particle volume concentration derived from the total aerosol particle number size distributions under the assumption of spherical particles (see subsection 2.3). Normalisation enables comparison of INP properties across different studies, which can include different approaches to INP number derivation, independent of* N_{INP}*. The resulting spectra of ice-active number site density,* n_s*, and volume site density,* v_s*, are given in Fig. S8b,c. Values of* n_s *spread over 4 orders of magnitude (10□¹–10³ cm□²) in the observed temperature range. For comparison, values from two laboratory experiments are included in Fig. S8b, that focused on sampling of artificially generated SSA and assessing its ice-activity (DeMott et al., 2016; Mitts et al., 2021). The results from DeMott et al. (2016) span a wider range of* n_s *and* T *than the ACE data, while values from Mitts et al. (2021) overlap with the open ocean-sampled filters from ACE. Field measurements from CAPRICORN-I (McCluskey et al., 2018a) showing a large variability in* n_s*, are included for comparison. Contrary to* N_{INP} *(Fig. S8a),* n_s *values from ACE lie within the lower end of what is reported from CAPRICORN-I. This indicates observation of a similar or more ice-active particle population during CAPRICORN-I compared to ACE. The range of* v_s *reported in Mitts et al. (2021) are included in Fig. S8c for comparison. The range overlaps with the lower range of the values from ACE. In conclusion, the strong overlap between ice-active site density profiles from ACE (derived from* N_{INP}*) and studies of artificial SSA (e.g., DeMott et al., 2016; Mitts et al., 2021) supports the idea that low* N_{INP} *measured on the open ocean might be driven by SSA."*

L736 tracked changes: *"A number of additional INP parameterisations are available in the literature that include the normalisation of the INP concentration to the particle surface (e.g., McCluskey et al., 2018a) or volume (e.g., Mitts et al., 2021). Both approaches of normalisation were performed with the ACE data (Fig. S8b,c) and show good agreement with previous studies of global marine environments. However, as discussed below the absence of a correlation between* N_{INP} *and neither* PM_{10} *nor* N_{total} *shows that both denominators (particle volume or surface) for the normalisation are directly linked to the INP."*

References: Mitts et al. (2021), doi: 10.1029/2020GL089633

RC2.5: Why are values above or below the INP detection limit included in figures and analysis? I find this very misleading and distracting. By including the below (above) detection limit values, the impression will be that the INP number concentrations are higher (lower) than what they may actually be. Because you are unable to measure, these values should simple be excluded (NaN).

AC2.5: There are two options: 1) to exclude the values on the border of the detectable range or 2) to include these values. To our understanding, both approaches cause a bias in the averaging of the considered values. We addressed this issue in the text (L480–486, L612–620 tracked changes) and provide average values for both cases in Tab. S2. This gives the potential user the option to choose the best version of the data set fit for purpose.

Introduction

RC2.6: Lines 83-85 - I do not believe the CAPRICORN II INP measurements have been publish other than in the McFarquhar et al. (2021) overview paper; McCluskey et al. (2018) only includes CAPRICORN I. (see also Lines 90-92)

AC2.6: Thank you, this is correct. The text has been changed accordingly, to reflect that McCluskey et al. (2018a) reported the CAPRICORN I cruise and data from CAPRICORN II is presented in McFarquhar et al. (2021).

L83 (L88 tracked changes): *"Two recent cruises: the Cloud, Aerosols, Precipitation, Radiation and Atmospheric Composition campaign (CAPRICORN-I & II). For CAPRICORN-I, observed* N_{INP} *over the SO in the temperature range between –12 and –31°C varying between 0.04 and 1000 m□³ (McCluskey et al., 2018a)."*

L91 tracked changes: *"Preliminary INP results for CAPRICORN-II are presented in McFarquhar et al. (2021) and underline the findings for CAPRICORN-I of low but highly variable* N_{INP} *values on the SO."*

RC2.7: Note that Bigg also hypothesized a decade decline in southern ocean INP concentrations (see Bigg 1990).

AC2.7: The reference has been included as hypothesis in addition to the differences arising from the different methods used to derive INP number concentrations (L493 tracked changes).

"Further, the highest concentrations were found near land and values differed largely from historical measurements (e.g., Bigg, 1973). Feedback of the Earth's changing climate on INP in the SO region as a contributor to the observed difference between current and historical observations cannot be ruled out (e.g., Bigg, 1990). However, potential mechanisms behind such a hypothetical feedback have not been identified either, tot he best of our knowledge."

RC2.8: Line 95: "In consequence, a dominance of sea spray on INP was concluded." - this should specify that the Southern Ocean *marine boundary layer* was dominated by sea spray INPs.

AC2.8: The sentence has been edited as suggested (L95, L102 tracked changes).

"In consequence, a dominance of sea spray on the INP population in the SO's MBL was concluded."

RC2.9: Figure 1 - Can the locations of the key ports and locations be added for

the reader to more easily follow along?

AC2.9: Fig. 1 has been updated to include markers and labels indicating the locations of Cape Town, Hobart, and Punta Arenas (P4, P5 tracked changes).

Methods

RC2.10: What size range is measured with the CCN instrument and how does this compare to the PM10 ionic composition measurements used in the correlation analysis? Can the authors provide some insight into the size ranges when introducing the correlation analysis in Section 3.4?

AC2.10: The CCN counter sampled ambient air through a PM_{40} inlet (shared by multiple instruments in the measurement container) and the HV sampled ambient air through a PM_{10} inlet (sampler-exclusive). The usage of these particular inlets defines the upper limit of the sampled particle size range to 40 µm and 10 µm, respectively. With the help of the size distributions, we calculated the respective critical dry diameter to the CCN concentration value (subsection 2.2) and found, on average, values between 30 and 100 nm (Tab. S1), depending on the *SS*. These critical dry diameter values are the lower limit of the particle size range sampled by the CCNc. The lower limit of the HV sampler depends on the filter material. In our case, filters consist of quartz fibres (MK 360 by Munktell) and have a particle retention of 99.9995% at around 300 nm (according to test method ASTM D 2986-91). However, giving the sampled size range is less important than the fact that number and mass dominated quantities are correlated. This is addressed in the text (L575–577, L717–719 tracked changes).

RC2.11: Line 180 - "sampling time (<1 to 1437 min) dependent on the automatic shut-down mechanism." - are the sampling volumes available or perhaps a mean/median sampling volume with standard deviation/IQR would help inform the reader of the typical sampling volume, since this statement suggests the sampling time ranged from 1 minute to ~24 hours.

AC2.11: The mean and one standard deviation of the sampled volume have been added for clarity (L179, L201 tracked changes).

"Here, filters showed an average sampled volume of 471.3 ± 151.4 m³ (mean ± SD) due to individual sampling times (<1 to 1437 min) depending on the automatic shut-down mechanism."

RC2.12: Lines 176 - was the automatic shut-down mechanisms used for both LV and HV? Please clarify.

AC2.12: Indeed, the text was not clear enough that the automatic shut-down mechanism was only used for the HV sampling and has been changed accordingly (L116, L138 tracked changes).

"An ultrasonic anemometer was operated next to the high-volume filter sampler and provided wind direction data for an automatic shut-down mechanism exclusive to the high-volume sampler."

RC2.13: Line 233 - should this be "specific depths down to 200 m" ?

AC2.13: The formulation has been changed as suggested (L232, L264 tracked changes).

"During the ACE cruise sea water was sampled every four hours using the RV's underway water supply system and during CTD (conductivity, temperature and depth) rosette deployments, at specific depths down to 200 m (Walton and Thomas, 2018)."

Results

RC2.14: Figure 2 - The caption mentions data were removed for instrument availability and ship exhaust filtering. There are many instances where CCN data are available, but the size distribution measurements are missing; are all of these times when the SMPS & APS were down?

AC2.14: Yes. The same data filter for the ship exhaust was applied to the CCN data as for the SMPS and APS data. Meanwhile, the instrument availability for the CCN instrumentation is independent of the availability of the SMPS and APS instruments.

RC2.15: Figure 3 - It's quite challenging to see the differ points. Suggest to widen the size of this figure such that the space between the supersaturation bins are wider?

AC2.15: Fig. 3 has been updated to address this issue and improve readability (P11, P13 tracked changes).

RC2.16: Line 296 - ".. cancel each other out so that no clear latitudinal" - should this be longitudinal?

AC2.16: Yes, please see AC1.16.

RC2.17: Line 307 - Can the authors expand on the second reason for differences (number of measurements during Austral summer is higher (three years)? Why would this lead one to expect differences?

AC2.17: Thank you for your question. We were referring to a potential effect of the inter-annual variability. However, we cannot think of good reasons actually. It might be the location of the station and the methodology. The text has been re-formulated accordingly (L306, L353 tracked changes).

"A first hypothetical reason for the differences at SS = 0.2 % is that PES is not located directly at the Antarctic coast."

RC2.18: Line 308 - I found the wording of this confusing; suggest change to "Disagreement of CCN concentrations at lower SS between Herenz et al. (2019) and this study suggests that differences arise from larger particles that are typical of activation at lower SS" ?

AC2.18: The sentence has been changed as suggested (L307, L355 tracked changes).

"A second hypothetical reason is that activation at this low SS *is associated with large particles, which might be removed due to atmospheric processes during transport to PES."*

RC2.19: Line 310-316 - I think the authors are trying to describe two sets of data that are included in Figure 3, but I think the labels may be inconsistent with the text - The "all data" I think refers to the first portion of this discussion and the "baseline" data refers to the later. However, the text refers to these as "baseline" (Line 313) and then mentions baseline values removed but reference the triangle right (line 316), which is labelled as "CGBS baseline" in Figure 3. Can the authors clarify?

AC2.19: The wording was not specific enough to emphasize that the later includes only measurements during "baseline" conditions. The text has been re-phrased accordingly (L315, L365 tracked changes).

"Averaging CGBS measurements which feature only baseline conditions, a median of ~130 cm³ (triangle pointing right in Fig. 3a) was found."

RC2.20: Line 317 - "We conclude that terrestrial influence on our average values is small" should specify average values of *CCN_0.5*, since data are unavailable for other SS.

AC2.20: "$N_{CCN,0.5}$" has been added for clarity as suggested (L316, L366 tracked changes).

"This is at the lower end of our results for Leg 1 and we conclude that the terrestrial influence on our $N_{CCN,0.5}$ *average values is small."*

RC2.21: Line 322 – suggest to include definition of BSO again here.

AC2.21: The text has been updated accordingly (L321, L373 tracked changes).

"For the British Southern Ocean (BSO) cruise, only CCN concentrations inferred from nss-sulfate are available in O'Dowd et al. (1997), not comparable with any of our N_{CCN}*."*

RC2.22: Lines 300-330 - I appreciate the thorough review of previous measurements - I do find the discussion challenging to follow with the different seasons, years, SS, etc. I suggest adding a table that summarizes these aspects. Also, are all of these studies evaluating the same aerosol size range?

AC2.22: Reference values were summarized in Tab. 1. Generally, we only considered studies for comparison, if the cut-off (PM_{40}) and CCN instrumentation were comparable to our measurements. However, if not, then it is mentioned in the text (e.g., L357).

RC2.23: Figure 4 - Should the hygroscopicity parameter values be on a linear

scale (as in Figure 3B)? Also, could points be added to these PDFs (and to Figure 7) to provide an idea of the bin sizes used?

AC2.23: The log-scale is used since the κ formula from Petters and Kreidenweis (2007) is a function of D_{crit} and the logarithm of SS. Fig. 4 clearly shows that our derived κ values follow a logarithmic distribution. Markers have been added to the figure to make the bins more visible. Note, there are 10 bins per order of magnitude, 30 in total.

RC2.24: Figure 5 – Are these data from the LV or HV filters? Please clarify. (same for Figures 6&7)

AC2.24: The captions for Fig. 5, Fig. 6, and Fig. 8 have been updated to clarify (see below).

Fig. 5: *"Time series of INP number concentration (N_{INP}) at (a) –24, (b) –20, (c) –16, (d) –12, and (e) –8°C from the LV filters sampled during ACE."*

Fig. 6: *"Mean values (crosses) and box-and-whiskers plots indicating the median (horizontal lines), inter-quartile range (boxes), and 10^{th} to 90^{th} percentiles (whiskers) of INP number concentration (N_{INP}) from the LV filters sampled during Leg 1 (green), Leg 2 (orange), and Leg 3 (purple)."*

Fig. 8: *"Normalized probability density functions (solid) and geometric mean values (dotted) for INP number concentrations (N_{INP}) at (a) –24, (b) –20, (c) –16, (d) –12 and (e) –8°C from the LV filters sampled during ACE."*

RC2.25: Line 398 - Values above the upper detection limit should be excluded, see Major Comment 4.

AC2.25: Please see AC2.5

RC2.26: Line 403 - "assessment of air mass origin is not possible for ACE" - See Major Comment 2.

AC2.26: Please see AC2.3

RC2.27: Line 411 - "the derivation of N_INP was fundamentally different" - how was it different?

AC2.27: Unlike our method of N_{INP} derivation, which utilizes a freezing array method, Bigg (1973) used a thermal diffusion chamber. The text has been changed accordingly (L411, L477 tracked changes).

"The techniques to measure N_{INP} were however different. Filter sampled during ACE were analysed with a freezing array method (subsection 2.3), while INP contents in Bigg (1973) were analysed by means of a thermal diffusion chamber."

RC2.28: Lines 411-415 - McCluskey et al. (2018) also discussed the hypothesis put forth by Bigg (1990) that a decadal decline in N_INP was possible over the Southern Ocean. INP concentrations reported in McCluskey et al. (2018) are also consistent with more recent measurements (McFarquar et al. 2021); It is not clear to me why so much focus is on comparing to Bigg (1973) without also considering more recent data. See Major Comment 1.

AC2.28: Please see AC2.2

RC2.29: Line 419 - See Major Comment 4.

AC2.29: Please see AC2.5

RC2.30: Line 440 - The Tobo et al. (2013) study was specific to terrestrial primary biological aerosol particles (PBAP) and I am unaware of marine INP studies that have identified a relationship between PBAP and INPs in a marine environment.

AC2.30: We agree that Tobo et al. (2013) focuses on particles of terrestrial origin. Studies showed that biogenic particle sampled in the Arctic act as INP at temperatures above ca. -16°C (McCluskey et al., 2018b; Hartmann et al., 2021). We assume that the same holds true for the Southern hemisphere. The text has been updated accordingly (L440, L546 tracked changes).

"PBAP were found to act as INP in several studies in marine regions of the Northern hemisphere (e.g., McCluskey et al., 2018b; Hartmann et al., 2021) and we assume the same to be the case for the Southern hemisphere. Therefore, we conclude PBAP from SSA to be a potential source for the INP measured on the open ocean sections of the cruise."

References: McCluskey et al. (2018b), doi: 10.1029/2017JD028033; Hartmann et al. (2021), doi: 10.5194/acp-21-11613-2021

RC2.31: Line 459 - "In other words, the PDF given is representative for the marine environment" I do not think the analysis supports this statement. It is also confusing to follow this up with "Note that INP active at this temperature range can be either of mineral nature and long-range transported from terrestrial sources, or originating from marine sources."

AC2.31: Following RC1.5, we realised the interpretation of $N_{INP,-24}$ is not helpful, as values are close to background levels indicated by the FBF. Consequently, the passage, as well as the follow-up, have been removed (L455, L565 in tracked changes).

RC2.32: Figure 8 - Are all samples included here, even ones with expected terrestrial influence from the ports (Grey shaded periods from Figure 5)? Suggest to segregate data using the land proximity data (i.e., data within grey shade versus not shaded from Figure 5).

AC2.32: Yes, samples from all parts of the cruise are included in the correlation analysis

(subsection 2.5). Following the suggestion, we performed the correlation analysis with only a subset of our data set. We used a threshold in $N_{INP,-16}$ (≤ 10 m□³) to assess the correlation for marine background INP levels, consistent with our findings on the air-mass origin during low $N_{INP,-16}$ cases (Fig. 7). This subset (n = 75) showed only slight differences compared to the full data set (n = 89) in terms of ρ values. For example, between $N_{INP,-12}$ and $N_{INP,-20}$, a ρ of 0.59 was found, which is lower than the ρ of 0.73 for the full data set. Lower correlation coefficients for this subset were also found for N_{total} and $N_{CCN,1.0}$ (0.59 compared to 0.62) and N_{mode2} and $N_{CCN,0.5}$ (0.599 compared to 0.61). These 3 differences in ρ are very close to our threshold for a meaningful correlation of 0.6. For the subset associated with terrestrial/coastal influence ($N_{INP,-16}$ > 10 m□³), the correlation analysis could not be performed due to the low number of samples (n = 11) for a number of quantities. However, this does not contradict the assumption that our analysis is representative for marine background conditions.

RC2.33: Line 479 - Note that a similar range of INP number concentrations is reported in McFarquhar et al. (2021) that expands the seasonal and spatial representation.

AC2.33: Please see AC1.4

RC2.34: Section 3.2 - I find it unclear if the authors think INP observations are thought to be representative of "open ocean" or were influenced by terrestrial air masses, or both throughout this discussion. I think it's both (based on abstract), but overall I suggest reorganizing this to have a clear consistent message for the reader.

AC2.34: We find that our INP observations contain both signals from the open ocean and terrestrial/coastal sources. We hope, the inclusion of the analysis of the air-mass origin (AC2.3) helps to make things clearer.

RC2.35: Line 560 - What wind speeds were typical? I do not see windspeeds included anywhere. Is the relationship between sea salt or SSA and windspeed expected to be linear?

AC2.35: The cited literature shows that the relation between wind speed and sodium concentrations is not necessarily linear, but logarithmic. Typical values for U_{10} during ACE have been added for reference. The text has been edited, to account for both (see below).

L556 (L692 tracked changes): *"Other studies in the SO region found positive but non-linear connections between wind speed and sodium mass concentrations (e.g., Schmale et al., 2013; Yan et al., 2020a; Landwehr et al., 2021)."*

L559 (L696 tracked changes): "*Note that the wind conditions encountered during ACE are characterised by median values of 9.88 (Leg 1), 6.62 (Leg 2), and 8.85 m s□¹ (Leg 3; Schmale et al., 2019) and the relative variability of the daily* U_{10} *averages is roughly ±70 %."*

RC2.36: Line 565 - Can the authors clarify how the PM10 measured size range

compares to the CCN measured size range (from Line 345: "For the size range between roughly 37 and 123 nm probed with our SS (Tab S1)") and what this would mean for comparing mass concentrations from the full size range from the HV filters?

AC2.36: Please see AC2.10

RC2.37: Line 591 - What is meant by a "delayed connection"?

AC2.37: The sentence has been extended to clarify how INP abundance could respond to Chl-*a* signals in a time-shifted manner (L590, L732 tracked changes).

"This suggests that the measured INP are not originating from local biogenic sources but does not exclude a time-shifted response."

RC2.38: Lines 591-598 - Are the authors aware of the existing marine INP parameterizations based on total aerosol surface area (McCluskey et al., 2018) aerosol volume (Mitts et al., 2021) - see Major Comment 3. Aerosol type-specific parameterizations may provide a lot of insight as to the INP type.

AC2.38: Please see AC2.4

Conclusions

RC2.39: Line 632 - "INP spectra (Fig 8) for the most part of the cruise feature similar levels and temperature range between -12 and -24degC." - What is meant by "similar levels"? I see 3 orders of magnitude variability in N_INP for any given temperature between -20 and -7degC in Figure 8. From the abstract (Lines 12-13) "[INP] concentrations spanned up to 3 order of magnitude, e.g., at -16degC from 0.2 to 100 m-3.".

AC2.39: We refer to the spectra showing steadily increasing N_{INP} with decreasing temperature for almost every filter. This is in contrast to the feature of strong increases N_{INP} within a small temperature range mentioned later (see below).

L588 tracked changes: *"A typical, steady increase in* N_{INP} *with decreasing temperature (1 order of magnitude per 5K) can be observed for the majority of filters and the FBF's curve (pink line)."*

L632 (L785 tracked changes): *"INP spectra (Fig. S8a) for the most part of the cruise feature a steady increase in INP concentration with decreasing temperature."*

RC2.40: Lines 634 - "We interpret this signal [correlations between N_INP in the temperature range between -12 and -24degC] as indication that mixed long-range-transported populations of INPs of biogenic origin (T>-20 deg C) and mineral dust (T <. -20degC) were present. " - Again, it is not clear to me that this statement is supported by evidence available in this study.

AC2.40: As evidence for the sampling of biological INP, we can only point to several filters showing features in the spectra of increased concentrations at high temperatures ("bio

bumps”). Since there was no thermal-treatment-check performed with the filters, there is no proof for the sampling of biological particles acting as INP. The following text has been added to emphasise on the hints we see regarding the sampling of particles of biological origin acting as INP (see below).

L589 tracked changes: *"At temperatures above −20°C, a feature of sudden increase in curve steepness for a number of filters is apparent. This feature is unlike the previously mentioned, steady increase in* N_{INP}*. A high concentration of INP above −15°C is typically associated with a signal of a biological INP source mixed with a mineral or less efficient INP source (e.g., Creamean et al., 2019)."*

L786 tracked changes: *"Features in the spectra of increased* N_{INP} *at warm temperatures indicate warm-temperature INP which are connected to biological origin (e.g., Creamean et al., 2019)."*

References: Creamean et al. (2019), doi: 10.5194/acp-19-8123-2019

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2021-700/acp-2021-700-AC2-supplement.pdf