RC = Reviewer Comment, AC = Author Comment, new suggested text in blue

Note, line numbers refer to the original manuscript, the location in the revised can be seen best in the tracked changes version document. We indicate these line numbers in ().

Answers to Reviewer 1

Anonymous referee #1, 01 Apr 2022

Summary

RC1.1: The authors have satisfactorily addressed the main comments from my initial review. The manuscript now includes a better introduction with reference to the studies of Schmale et al. (2019) and Welti et al. (2020), the comparison with McCluskey et al. (2018) has received more attention, and the discussion of the PDFs has also been greatly improved by linking them to trajectory results shown in Fig. 7. I still found a number of mostly technical corrections, the list below may not be complete, and I would encourage further careful proofreading.

AC1.1: We thank the reviewer for the positive review. The proposed technical corrections have been addressed.

RC1.2: P2L43/44: "… it is called a cloud condensation nucleus."

AC1.2: The sentence has been edited as suggested (L44, L44 tracked changes).

Particles acting as nuclei for cloud droplet formation at atmospherically relevant (water vapour) supersaturation are commonly referred to as cloud condensation nuclei (CCN).

RC1.3: P3L85: The sentence starting with "Two recent …" is not complete.

AC1.3: The sentence has been changed in order to follow a complete grammatical structure (L86, L87 tracked changes).

Two recent cruises were conducted as part of the Cloud, Aerosols, Precipitation, Radiation and Atmospheric Composition campaign (CAPRICORN-I & II).

RC1.4: P3L87: It should be "varied" instead of "varying"

AC1.4: The sentence has been edited as suggested (L87, L88 tracked changes).

For CAPRICORN-I, observed N_{INP} over the SO in the temperature range between -12 and -31°C varied between 0.04 and 1000 m^{-3} (McCluskey et al., 2018a).

RC1.5: P9L246: Maybe: "Integration gives the total particle surface area and volume for each measurement."

AC1.5: This part, including the suggested sentence, has been updated (L245, L246 tracked changes).

For this, the particle surface area and volume size distributions were first inferred from the number size distribution, assuming spherical particles, and then integrated over the entire diameter range. This was done for each size distribution measurement.

RC1.6: P9L248: I am bit wondering about the variable name “v_s”. I see that it is also used in Mitts et al. (2021), but wouldn’t the names “n_s” for the surface density of active sites, and “n_v” for the volume density of active sites be more appropriate? The same applies to the labelling of the y-axes in Figs. 8b and c (which again is the same as in Mitts et al.), why is “n_s” referred to as number site density and “v_s” as volume site density? I would call them “surface density of active sites” or “number of active sites per unit surface area of INPs” respectively “volume density of active sites” or “number of active sites per unit INP volume”. But please correct me if I have a misunderstanding here.

AC1.6: We agree that n_v (instead of v_s) as the symbol for the volume site density is a better choice. The text and Fig. S8 have been changed accordingly (see below).

- L247 (L250 tracked changes): These values were averaged over the 8 h sampling time of each LV filter and N_{INP} is divided by these values, resulting in the ice active site density (n_s) and ice active volume density (n_v), respectively.
- L548 (L556 tracked changes): The resulting spectra of ice-active number site density, n_s, and volume site density, n_v, are given in Fig. S8b,c.
- L556 (L563 tracked changes): The range of n_v reported in Mitts et al. (2021) are included in Fig. S8c for comparison.
- L10 in the SI: Figure S8 shows the temperature-dependence of N_{INP}, n_s, and n_v for the LV filters.
- Fig. S8 caption (P12 in the SI): Temperature-dependence of (a) INP number concentration N_{INP}, (b) number site density n_s, and (c) volume site density n_v for the LV filters sampled during ACE. Values of n_s (n_v) were calculated by normalising N_{INP} with the total particle surface area (total particle volume) derived from an averaged particle number size distribution per filter under the assumption of a population of only spherical particles.

RC1.7: P13L351: Incomplete sentence. Maybe: “The averaging of the CGBS measurements … gives a median of …” And delete “was found” at the end.

AC1.7: The sentence has been revised to make it complete (L353, L356 tracked changes).

The averaging of the CGBS measurements which feature only baseline conditions gives a median of ~130 cm^{-3} (triangle pointing right in Fig. 3a).

RC1.8: P14L361: “are reported in Sanchez et al.”

AC1.8: The sentence has been edited as suggested (L362, L365 tracked changes).

As for aircraft-based CCN measurements, $N_{CCN,0.3}$ between 17 and 264 cm^{-3}, with an average of 123 ± 58 cm^{-3} (mean ± SD), are reported in Sanchez et al. (2021) for flights through the MBL between 42.5–62.1° S and 133.8–163.1° E during the Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES).

RC1.9: P14, header of Table 1: Duplicate header label “Time frame”

AC1.9: Tab. 1 has been changed accordingly.

RC1.10: P17L431: Sentence is also difficult to read, maybe: “INP concentrations outside the detectable range (indicated by triangles and estimated as described in subsection 2.3) were included

in the figure as they mark …" Why do the maximum INP concentrations represent the lower detection limit and vice versa?

AC1.10: We agree that this sentence needs to be improved in terms of readability. As for the representation of the detectable range of N_{INP}, the text was misleading. A value outside the detectable range is represented by the maximum (minimum) value of the lower (higher), unresolvable concentration range. The sentence has been edited as suggested and the idea behind giving the detection limit has been clarified (L433, L437 tracked changes).

INP concentrations outside the detectable range (indicated by triangles and estimated as described in subsection 2.3) are represented in the figure by each filter's lowest (lower detection limit) and highest resolvable concentration value (upper detection limit).

RC1.11: P19L462: Why does the exception also apply to -20°C? For -16°C it is clear to me (Fig. 5c), as your measurements on the open ocean nicely fall into the range of the McCluskey data, but for -20°C (panel b) it seems to me that they are also at the upper end of the CAPRICORN-I data.

AC1.11: We agree that this is not the case at -20°C. The sentence has been edited accordingly (L464, L469 tracked changes).

At each temperature, N_{INP} observed during ACE are at the upper end or higher than concentrations observed during CAPRICORN-I, except at -16°C when low concentrations were measured on the open ocean in air-masses without terrestrial influence.

RC1.12: P19L470/471: Maybe: "have not been identified so far"

AC1.12: The sentence has been edited as suggested (L473, L478 tracked changes).

However, potential mechanisms behind such a hypothetical feedback have not been identified so far.

RC1.13: P22L535/536: The description "a feature of sudden increase in curve steepness" was not particularly clear to me, perhaps it would be better to speak of a shoulder in the INP spectrum at higher temperatures, which could indicate biological INP sources.

AC1.13: We agree that "shoulder" is a simpler description of the feature commonly known as "bio-bump". This part of the text has been updated accordingly (L539, L545 tracked changes).

At higher temperatures (>-20°C) a shoulder in the INP spectra of a number of filters is apparent.

RC1.14: P23L544/545: Please explain more clearly what you mean by "which can include different approaches to INP number derivation, independent of N_INP". As I understand it, n_s would just be a concept to normalize the results of studies where different particle sizes were used.

AC1.14: We agree that the text was not clear enough on why the normalisation is used. The sentence has been edited as suggested (L547, L554 tracked changes).

Normalisation enables comparison of INP properties across different studies, which can include different N_{INP} derivation approaches or sampled particle size ranges, independent of N_{INP}.

RC1.15: P23L558: Better phrase it like e.g.: "For the sake of completeness, however, we also show

in Fig. S5 the additional INP spectra determined from the HV samples (…).”

AC1.15: The sentence has been edited as suggested (L561, L569 tracked changes).

For the sake of completeness we give in Fig. S5 the additional INP spectra determined from HV samples (*DHA-80* sampler, see subsection 2.3).

RC1.16: P26L641: Repetitive “The lack of a correlation”. Maybe just state: “This might be a smearing effect …”

AC1.16: The sentence has been edited as suggested (L646, L654 tracked changes).

This might be a smearing effect due to averaging, since the highest N_{CCN} (subsection 3.1) coincide with the highest M_{MSA} (subsection 3.3) when the cruise was in the vicinity of the Antarctic coast (Leg 2).

RC1.17: P27L652: You added “that” after “supports” in the revised version, but I don’t think this is correct.

AC1.17: The sentence has been edited as suggested (L657, L665 tracked changes).

This again supports the observation of SSA particles not significantly contributing to the CCN population, as SSA dominates the sampled particle mass but not the particle number.

RC1.18: P29L713: “The comparison with other studies shows that the …”

AC1.18: The sentence has been edited as suggested (L718, L726 tracked changes).

The comparison with other studies shows that the concentrations observed during ACE are lower than what was observed previously over the SO (Bigg, 1973), while being on the upper end of what is reported in McCluskey et al. (2018a) for a specific sector of the SO in March–April 2016 (Fig. 5 and Fig. S8a).

Answers to Reviewer 2

Anonymous referee #2, 06 Apr 2022

Summary

RC2.1: In the paper submitted titled “Circum-Antarctic abundance and properties of CCN and INP”, Tatzelt et al. describe results from the 2016/2017 research voyage that aimed to characterize southern ocean CCN and INPs. The authors have addressed several comments, including an impressive back trajectory analysis that really add to the discussion. Below are several minor comments and one major comment that I will provide related to the inclusion of background INP number concentrations.

AC2.1: We thank the reviewer for the positive and constructive review. The inclusion of the field blank filter INP number concentrations was carefully taken into consideration. We have made changes to the text following the major and general comments below.

Major comment:

RC2.2: Lines 237-239: “Based on the fice of the field blank filters (FBF) we determined averaged temperature-dependent INP concentrations, NINP;FBF, which are given as point of reference for background concentration levels whenever NINP for the sampled filters are shown.” -yet, the FBF results are only shown in the supplemental figures (Figures S5 and S8) and are not included in figures 5, 6, or 8. I do not think adding the FBF to these figures will significantly change the results of the paper. However, not including these FBF results carefully could lead to mis-use of the data if someone were to use these data in future comparisons to other observations or model estimates in the future. Given that many of the measured values reported are below the background levels measured from the FBF, I think this is important to address and suspect it will also be straight-forward to add.

AC2.2: We agree that the addition of the FBF-derived N_{INP} in the INP figures gives context to the presented, encountered values. Consequently, the mean $N_{INP,FBF}$ has been added to Fig. 5 and Fig. 6 as suggested and the respective captions have been updated (see below). As for Fig. 8 (PDF of N_{INP}), we decided against the inclusion of the mean $N_{INP,FBF}$ due to a poor readability of the resulting figure. We hope, the message that our values are at times close to the background comes across in the other two figures regardless.

- Fig. 5 caption (P18): In (a–d) the measurement background from averaged spectra of field blank filters (FBF) is indicated (dash-dotted lines).
- L435 (L438 tracked changes): For $T \leq$ -12°C (Fig. 5a—d), the respective measurement background INP concentrations are represented via the averaged FBF spectra (dash-dotted lines), as described in subsection 2.3.
- Fig. 6 caption (P20): In the figure, the measurement background is represented by the averaged spectra of the field blank filters (FBF; dash-dotted lines) and the number of data points (n) are indicated.
- L484 (L491 tracked changes): As a point of reference for the measurement background, concentrations of the averaged FBF are included (dash-dotted lines).

Typos/minor comments:

RC2.3: L84 – It might be helpful to include additional information on the freezing mode of focus of this work.

AC2.3: We agree that immersion freezing, the freezing process used to investigate the sampled INP, needs direct statement in the text. Information on the used freezing method has been added the abstract (L11, L11 tracked changes).

Ambient INP number concentrations were measured in the temperature range from -5 to -27°C using an immersion freezing method.

RC2.4: L85-86 – “two recent cruises:…” is not a complete sentence.

AC2.4: Please see AC1.3

RC2.5: L94 – “… whether sampled INP have terrestrial or oceanic sources..” I think the word “have” should be replaced with the word “had”

AC2.5: The sentence has been edited as suggested (L95, L96 tracked changes).

Correlation of INP and ambient radon concentration was used to assess whether sampled INP had terrestrial or oceanic sources for the CAPRICORN-I cruise.

RC2.6: L134 – ".. wind direction within a 180deg half-circle at the sampler, with the RV's exhaust at 90deg" – I find this very challenging to follow. I think you mean that the exhaust was behind the sampler and the 180deg half-circle is in front the sampler? Can you please clarify?

AC2.6: We agree that description of the exclusion wind directions was not straightforward. The sentence has been edited to improve understandability (L135, L136 tracked changes).

Sampling was stopped automatically during periods with wind direction within a half-circle at the sampler centred towards the stack exhaust situated on the RV's stern.

RC2.7: L240/Table S3 – why are the HV FBF results not included in Table S3 for comparison?

AC2.7: We agree that the addition of the FBF INP concentrations for the HV samples will give context to the values presented, e.g. in Tab. S4. These values have been added to Tab. S3 and the respective caption has been updated accordingly.

- L241 (L242 tracked changes): In Tab. S3 $N_{INP,FBF}$ is given for the LV and the HV filters.
- Tab. S3 caption (P14 in the SI): Mean INP number concentration of field blank filters (FBF) for LV sampling ($N_{INP,LV,FBF}$) and HV sampling ($N_{INP,HV,FBF}$) at selected temperatures (T).

RC2.8: L334 – "… considered representative for the whole SO region" – Important to mention that this is specific to the SO marine boundary layer and summertime. This statement might also fit better after the discussion of the previous CCN measurements?

AC2.8: We agree that the statement made in this sentence was too general. However, since the conclusions are drawn directly from what is shown in Fig. 3a, we think the sentence would not fit at an earlier point in the text. The sentence has been edited to include specifications on height and season (L335, L338 tracked changes).

With this, the CCN concentrations given in Tab. S1 can be considered representative for the MBL over the whole SO region during summertime.

RC2.9: L346 – "For this period, a median of ~230cm-3" – I think this referring to N_CCN,0.5, right? Can you please specify?

AC2.9: We agree that there is specification needed at this point. The sentence has been edited to indicate the *SS* of the N_{CCN} in question (L348, L351 tracked changes).

For this period, a $N_{CCN,0.5}$ median of ~230 cm^{-3} is reported (triangle pointing left in Fig. 3a).

RC2.10: Table 1 – note that the second column header (Time frame) has a typo in that it is redundant. Also, can the authors add if these studies were all summertime measurement campaigns in the caption?

AC2.10: The duplicate in the header of the second column has been removed (see AC1.9). Information on the common season of the presented studies has been added to the caption of Tab. 1 as suggested (P14, P14 tracked changes).

Overview of a selection of studies on aerosol particles and CCN over the summertime Southern Ocean.

RC2.11: L448 – "Contrary to these regions, air-masses passing over Antarctica did not show higher NINP than oceanic air-masses (Fig. S7)." – I recognize the authors are referring to Figure S7, but what about the period of elevated Antarctic airmass influence and elevated INP number concentrations between the January 26 and Feb 9 timestamps in Figure 5?

AC2.11: Looking into the data for Fig. S6d, we found elevated Antarctic air-mass influence (≥80%) for the period 28th January to 31st January 2017. This coincides with the RV being close to or stationed at Mertz Glacier (see cruise report in Walton & Thomas, 2018). The mentioned elevated INP concentrations (e.g. $N_{INP,-16}$>10 m^{-3} in Fig. 5c) are found for a sample from 2nd February 2017, when the RV was already on the Ross Sea. Further, Fig. S6c shows for 2nd February 2017 a strong influence of sea ice and the MIZ on the air-mass origin.

References: Walton & Thomas (2018), doi: 10.5281/zenodo.1443511

RC2.12: L453 – "Filter sampled during ACE …" should this be filters (plural?)

AC2.12: The sentence has been edited as suggested (L456, L461 tracked changes).

Filters 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.

RC2.13: L478 – "we report all averaged values…" – where are the average values for option A? Is that in Table S2?

AC2.13: We agree that the text was not clear enough on which approach led to N_{INP} (option A) and N_{INP}^{*} (option B) in Tab. S2. This part of the text and the caption of Tab. S2 have been updated to be more precise (see below).

- L476 (L482 tracked changes): Two different approaches were used for averaging the INP concentrations of the LV samples. In the first approach only values which are inside the detectable range are considered and the averages are given as $N_{INP,LV}$ in Tab. S2. For the second approach, values outside the detectable range ($N_{INP,LV}^{*}$) were included by using a value on the edge of the detectable range instead (see subsection 2.3). Results of the two approaches differ in mean, median, and geometric mean concentration values by up to ±50 %. The largest differences were found at a T of -8 and -24°C, where the number of data points outside the detectable range is largest.
- Tab. S2 caption (P13 in the SI): For this averaging only concentrations within the detectable range are considered and the number of samples, n, are indicated in the table.

RC2.14: L497 – ".. FBF concentrations are 0.08…" why were only the FBF concentrations mentioned for -12 and -16? The values for -20 and -24 deg C are 7.72 m-3 and 57.76 m-3 (Table S3), respectively, which falls very near the peak of the PDF in panels a and b. Please see major comment 1.

AC2.14: Please see AC2.2

RC2.15: L559-571 – somewhere in this discussion the authors should also discuss the higher background concentrations of the HV filters (Figure S5) compared to the LV filters (Table S3 and Figure S8). From eye, the FBF results associated with the HV filters are an order of magnitude

(factor of 2) higher than the LV filters for temperatures –12 and -16 deg C (-20 and -24 deg C), which would limit the ability to measure lower INP number concentrations, thereby making the spread more narrow and biased high. I think the existing discussion is also helpful, but this large difference in field blank background INP concentrations is likely important.

AC2.15: We agree that from the higher measurement background for the HV sampling one can assume the measured concentrations to be naturally higher. We included this idea in the text (L571, L580 tracked changes).

The higher background INP levels for the HV filters, indicated by higher $N_{INP,FBF}$ compared to the LV sampling (Tab. S3), hints on a limited ability of the HV sampling to measure lower INP number concentrations and in consequence overall higher measured INP number concentrations.

RC2.16: L581 – I think this should be Table S5.

AC2.16: The sentence has been edited as suggested (L585, L593 tracked changes).

Averaging sodium mass concentrations for the whole cruise gives a median value of 2.8 µg m^{-3}, with an IQR from 1.8 to 3.9 µg m^{-3} (Tab. S5).

RC2.17: Figure 7 caption: I think there is a typo, where the non-Antarctic land masses should be red, rather than light blue.

AC2.17: The caption of Fig. 7 has been edited as suggested (P21, P21 tracked changes).

Color codes for surface types are: non-Antarctic land masses (red), non-Antarctic coastal regions or islands (orange), Antarctic continent or coastal regions (ANT; yellow), ice-covered regions (light blue), and open ocean (dark blue).

Supplemental minor comments:

RC2.18: L15 – should the Thurnherr referenced be mentioned here?

AC2.18: The sentence has been edited as suggested (L15 in the SI).

Backward-trajectories for ACE are available in Thurnherr et al. (2020).

Reference: Thurnherr et al. (2020), doi: 10.5281/zenodo.4031705

RC2.19: L28 – “Here, the model’s land-sea mask” – which model?

AC2.19: The LAGRANTO tool uses ERA-interim reanalysis data which is the output of the ECMWF Integrated Forcasting System model. The text has been updated to include this information (see below).

- L15 in the SI: Backward-trajectories for ACE are available in Thurnherr et al. (2020). They used the Lagrangian analysis tool (LAGRATO) described in Sprenger and Wernli (2015) with reanalysis data from the European Centre for Medium Range Weather Forcasts (ECMWF). These reanalyses are produced by the ECMWF Integrated Forecasting System (IFS), an atmospheric model and data assimilation system.
- L31 in the SI: Here, the IFS's land-sea mask is used for initial classification of the surface signal type.

References: Sprenger and Wernli (2015), doi: 10.5194/gmd-8-2569-2015

RC2.20: L33 – “A commonly use sea ice fraction…” – is there a reference for this?

AC2.20: In our understanding, the commonly used approach of classifying grid boxes in atmospheric models as open ocean if the sea ice fraction of the grid box is below 15 % originates from Cavalieri et al. (1991) finding the best agreement for the sea ice edge location between airborne microwave radiometer observations and ship-based crossings for a sea ice concentration of 15%. The reference has been added to clarify the origin of the threshold value (L36 in the SI).

A commonly used sea ice fraction threshold of <15 % is applied here to classify as "open ocean" (following Cavalieri et al., 1991).

References: Cavalieri et al. (1991), doi: 10.1029/91JC02335

RC2.21: Figure S6 – Panel a is confusing to me. It is unclear how 10 hour trajectories would relate to “duration of trajectory cluster” values larger than 240 hours, and here the values go up to 1920 hours? I think I may be missing something.

AC2.21: For the airmass-origin analysis of the LV samples, the hourly clustering (and subsequent averaging) of all trajectories ending within the PBL was performed up to eight times, corresponding to the 8 h sampling interval of the LV filters. The maximum duration of 1920 h is reached in the case that each of the eight hourly time steps consists of a trajectory cluster with the maximum duration of 10 d (or 240 h). The text has been updated in an attempt to improve the explanation of the clustering procedure (see below).

- L19 in the SI: At an hourly resolution, trajectories are available for 56 pressure levels (between the surface and 500 hPa above sea level) above the RV's position. To achieve more robust statistics, for each hour all trajectories ending within the planetary boundary layer (PBL) above the RV's position are averaged into a cluster by calculating median values of latitude, longitude, pressure level, boundary layer pressure level, total precipitation, land fraction and sea ice fraction.
- L41 in the SI: The final step of the analysis is the averaging of PBL signals from all clusters within each LV filter's 8 h sampling period. The total number of trajectories that are averaged in clusters and considered per LV filter is between 1 and 28, with a mean of roughly 15.

RC2.22: Table S2 – I think additional details are needed on the first grouping of averages.

AC2.22: Please see AC2.13

RC2.23: Table S3 – As mentioned above, please add the HV FBF results. Also, should there be a range since more than 1 FBF was collected (L197 of main text) and a range of sampled volume was estimated for this calculation (L239-240)?

AC2.23: The $N_{INP,FBF}$ for the HV samples have been added to Tab. S3 (see AC2.7). We found the N_{INP} of individual FBF to vary between the range described by a factor 2 of the mean FBF. Given that the variability of V_{flow} is roughly 10%, we expect resulting variability for $N_{INP,FBF}$ in the same region. This is well within our conservative estimation for the uncertainty in FBF of a factor 2. With that we think it sufficient to show FBF ± 2 FBF in the spectra plots of Fig. S5 and Fig. S8.