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Reply on RC1

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.,
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Answers to Reviewer 1

Anonymous Referee #1, 25 Oct 2021

Summary

RC1.1: Tatzelt et al. present an extensive data set of CCN and INP measurements from the Antarctic Circumnavigation Expedition of 2016/2017. I understand this to be a very useful addition to the literature, as little data has been available for the Southern Ocean region so far. The article is well-written and contains well-elaborated figures that adequately convey the important features of the data set. The reader is well guided through the discussion, which is fairly detailed but avoids over-interpretation. I recommend publication in ACP, and only have a couple of specific comments and technical corrections as listed below.

AC1.1: We thank the reviewer for the positive and very constructive review. Their comments made our study more targeted, structured, and understandable. We have rewritten the text following the major and general comments below.

Specific comments

RC1.2: P4, L100-102: The introduction into the present study is rather brief. I think it would be fair to already mention here that parts of the data set have already been presented in Schmale et al. (2019) and Welti et al. (2020) (only mentioned later in line 416) and to summarize the main conclusions from these studies. Schmale et al. (2019) e.g. already presented some analysis on the role of sea spray for CCN. And then underline that you present here the full data set with all the CCN measurements at different supersaturation levels and the temperature-dependent INP records, as well as the correlation analysis to find potential links between the measured properties.

AC1.2: We now mention the overview paper for ACE and highlight the added value of the current study. The text was changed accordingly (L109 tracked changes).

"Parts of the CCN and INP data set presented in this study have previously been presented in the overview on the ACE cruise in Schmale et al. (2019). Aerosol properties were found

to be highly heterogeneous over the SO. The CCN abundance in the MBL showed a significant sea spray contribution in the strong westerly wind belt, while in the polynyas of the Ross and Amundsen Sea biogenic emissions are more important. INP abundance was shown to be lower on the SO than in Northern hemispheric marine air, with small differences between samples on the open ocean and close to the Antarctic coast. INP abundance was found to be similar to other studies on the SO (e.g., McCluskey et al., 2018a) but lower than historic data from Bigg (1973).

INP concentrations at -15°C from the ACE expedition have been presented in Welti et al. (2020). They show that N_{INP} from ship-based measurements are lowest in polar regions and highest in temperate climate zones. Overall, geographical variation in N_{INP} is below 2 orders of magnitude at any temperature. At low temperatures, lower N_{INP} were encountered in the Southern hemisphere than in the Northern hemisphere and this was attributed to the concentration of dust particles active as INP.

These two previous studies (Schmale et al., 2019; Welti et al., 2020) presented a subset of the CCN and the INP data in larger contexts. This paper focuses on the detailed analysis and interpretation of the observations, including N_{CCN} at all available SS and N_{INP} at the full investigated temperature range. Based on 10-day backward-trajectories, an air-mass analysis was performed to locate potential INP sources. In addition, a correlation analysis was performed using CCN, INP, and additional data from the ACE expedition in order to find potential links between the measured properties."

RC1.3: P9, L256: You may already include here a brief explanation that the LV filters allow for more robust statistics and were therefore included in the correlation analysis, and that also the overall interpretation of the INP results is mainly based on the LV filters. The HV-filter INP results are not mentioned for a long time (up to line 480) and you can state the reason here.

AC1.3: We inserted the following sentence to explain why the LV filter results were chosen for the analysis (L232 tracked changes).

"Due to the higher number of LV samples, resulting in more robust statistics compared to the HV samples, we focus in subsection 3.2 on INP results derived from the LV samples, while only briefly commenting on results from the HV samples."

RC1.4: P15, L408: Could you please also indicate the range of the McCluskey et al. (2018) INP results in Fig. 5 (similar to the Biggs data)? These data are referred to rather late and only very briefly in the discussion (L476 ff, Fig. 8). I think the comparison with McCluskey et al. (2018) deserves a more prominent place in the discussion already in the context of Fig. 5.

AC1.4: The data range of CAPRICORN-I (McCluskey et al., 2018a) has been added to Fig. 5. Further, the results for CAPRICORN-II and MARCUS (McFarquhar et al., 2021) have been added to the discussion (see below).

L486 tracked changes: *"In McCluskey et al. (2018a) INP measurements from CAPRICORN-I are presented. The range of observed INP concentrations is included in Fig. 5 for comparison. 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 and -20°C , when low concentrations were measured on the open ocean in air-masses without terrestrial influence. Differences in sampled geographical area (CAPRICORN-I: $43\text{--}53^{\circ}\text{S}$*

and 141–151 °E; this study: 34–78° S, circum-Antarctic) and season (CAPRICORN-I: March–April; this study: December–March) could be reasons for the differences in observed INP abundance."

L491 tracked changes: "Our results are consistent with preliminary results from MARCUS, CAPRICORN-I & II in McFarquhar et al. (2021), where N_{INP} in the MBL over the SO are shown to exhibit a large variability, very low overall values and a weak overall latitudinal dependence."

References: McFarquhar et al. (2021), doi: 10.1175/BAMS-D-20-0132.1

RC1.5: P16, L432: In particular for $N_{INP}(-16^{\circ}\text{C})$, the low concentration mode with 0.2 m^{-3} is not only close to the detectable range, but is also only a small addition on top of the mean INP concentration of the blank filters at that temperature (0.59 m^{-3} , Table S3). I doubt that it is justified to interpret it as an individual mode. Also the shape of the PDFs for -20 and -24°C could be strongly influenced by the background, as can be seen by the strong overlap of the measurement data with $\text{FBF} \pm 2 \cdot \text{FBF}$ in Fig. 8. This should at least be mentioned in the discussion.

AC1.5: The text has been expanded with the following part to emphasise that there is no point in over-interpreting the third, lowest concentration mode. In addition, the interpretation of -24°C was avoided due to the closeness to the FBF.

L524 tracked changes: "Interpretation of the PDF for $N_{INP,-8}$ and $N_{INP,-24}$ is omitted due to the low number of samples compared to other temperatures and concentrations being close to the FBF, respectively."

L431 (L529 tracked changes): "The lowest concentration mode for $N_{INP,-12}$ and $N_{INP,-16}$ include concentrations below 0.2 m^{-3} which are on the lower boundary of the detectable range. Attributing these concentrations to a source of geographical origin is ambiguous when considering the FBF as a point of reference for the background freezing signal. FBF concentrations are 0.08 and 0.59 m^{-3} for -12°C and -16°C , respectively (Tab. S3). We therefore only discuss the two highest concentration modes in the following."

Minor comments and technical corrections:

RC1.6: P1, L6: Add comma after "and".

AC1.6: The sentence has been edited as suggested (L5, L5 tracked changes).

"An overview of CCN and INP concentrations on the Southern Ocean is provided and, using additional quantities, insights regarding possible CCN and INP sources and origins are presented."

RC1.7: P1, L10: You frequently quote the numbers 37 – 123 nm for the average critical dry diameters in the text (also e.g. L345, L356), but the geometric mean values in Table S1 are rather from about 30 – 110 nm.

AC1.7: Thank you for pointing this out. The text has been updated for consistency (see

below).

L9 (L9 tracked changes): *"No distinct size-dependence of κ was apparent, indicating homogeneous composition across sizes (critical dry diameter on average between 30 and 110 nm)."*

L284 (L329 tracked changes): *"Consequently, quantities presented later in this manuscript, that are derived from $N_{CCN,1.0}$, are representative for the larger Aitken mode particles (D_{crit} at this SS \approx 30 nm, see Tab. S1)."*

L345 (L380 tracked changes): *"For the size range between roughly 30 and 110 nm probed by the range of SS (Tab. S1), the chemical composition appears to be independent of particle size, which further suggests a well-mixed aerosol (or CCN) population."*

L575 (L717 tracked changes): *"This indicates that the chemical information for CCN between 30 and 110 nm approximated by κ is not connected to the mass-dominating, seemingly larger particles represented by sodium and MSA."*

RC1.8: P2, L45: You may note here that this process is called immersion freezing, because you use this term later in line 82.

AC1.8: We now mention immersion freezing at this point of the manuscript (L189, L212 tracked changes).

"The immersion freezing capability of the aerosol particles collected on each LV and HV filter was measured using the Ice Nucleation Droplet Array (INDA) at TROPOS."

RC1.9: P4, L93: This is just one example where you sometimes switch from past to present tense when describing older studies, this should be homogenized.

AC1.9: The text was accordingly homogenized (L93, L100 tracked changes).

"Additionally, Uetake et al. (2020) showed that bacteria sampled during CAPRICORN-II are mostly of marine origin, suggesting a restricted meridional transport of continental aerosol towards the SO."

RC1.10: P5, L136: Maybe replace "Thereby" by "In this approach"

AC1.10: The text has been updated as suggested (L136, L158 tracked changes).

"In this approach, each PNSD is assumed to be a superposition of up to three aerosol modes and log-normal distributions in pre-defined size ranges are fitted."

RC1.11: P6, L142: Shouldn't it be "all geometric diameters"?

AC1.11: Thank you for your comment. Indeed, diameters have been converted from mobility to geometric diameter, as mentioned earlier in this paragraph. The text has been changed accordingly (L141, L163 tracked changes).

"Integration of smoothed total PNSD over all geometric diameters gave total aerosol particle number concentration (N_{total}) for each time step."

RC1.12: P10, second line of figure caption: dashed instead of dotted lines

AC1.12: The caption of Fig. 2 has been changed accordingly (P10, P11 tracked changes).

"Ports visited (dashed lines) and vicinity to land (grey area) are indicated in the figure."

RC1.13: P10, L287: There are occasional sentences with lots of insets and commas that are difficult to read. Here you could just delete both commas and also delete "individually".

AC1.13: The text has been revised in an attempt to simplify longer sentences. This sentence has been updated (L287, L332 tracked changes).

"Averages of $N_{CCN(SS)}$ for the Legs of ACE are shown in Fig. 3a."

RC1.14: P11, L292: As mentioned above, previous findings from Schmale et al. (2019) could also briefly be discussed in the introduction.

AC1.14: Please see AC1.2

RC1.15: Fig. 3: You could update Sanchez et al. (2020) to (2021) in the legend.

AC1.15: The reference in Fig. 3a has been updated accordingly.

RC1.16: P12, L296: You still discuss here that there is no clear longitudinal trend, don't you?

AC1.16: Yes, this sentence is dedicated to longitudinal observations. We changed the sentence to clarify this (L296, L340 tracked changes).

"The longitudinal differences in CCN abundance is either small against the overall variability in the data, or a variety of effects cancel each other out so that no clear longitudinal trend can be observed."

RC1.17: P13, L331: Delete the inset ", individually,".

AC1.17: The insert has been removed (L331, L383 tracked changes).

"An overview on the aerosol particle hygroscopicity parameter κ observed during Legs 1–3, is given in Fig. 3b."

RC1.18: P13, L343: Please homogenize the spelling of "Leg" with a first capital letter. (there are numerous instances)

AC1.18: We corrected the spelling of "leg" and "legs" to "Leg" and "Legs" throughout the text and figures.

RC1.19: P14, second line of figure caption: What is meant here by a "five minute resolution"? Does it refer to the measurement period per supersaturation set point?

AC1.19: We averaged 5 minute intervals of 1 Hz measurements. Each of the six SS-levels was run for 10 minutes and the first 5 minutes were discarded to ensure thermodynamic equilibrium in the CCNc. SS-settings were repeated once each hour. The caption of Fig. 4 has been changed to make this clearer (P14, P17 tracked changes).

"The N_{CCN} result from averaging 5 min long intervals of 1 Hz measurements. Each of the six SS-levels is repeated once per hour."

RC1.20: P15, L406/407: Please reformulate the sentence starting with "However ..." It is rather complicated. You could make a full stop after "land" and start a new sentence with, e.g.: "It does not account for the actual air mass origin and thus is not ..."

AC1.20: The part elaborating the methodology in Moallemi et al. (2021) has been removed (L404, L464 tracked changes).

RC1.21: P15, L409: The inset ", respectively" could also be deleted.

AC1.21: The insert has been omitted (L408, L475 tracked changes).

"They sampled filters in the SO around Australia, collecting 0.3 and 3 m³ of ambient air through a pair of membrane filters."

RC1.22: P16, L432: "exhibits" and "contains"

AC1.22: The sentence has been reformulated (L431, L529 tracked changes).

"The lowest concentration mode for $N_{INP,-12}$ and $N_{INP,-16}$ contains concentrations below 0.2 m³ which are on the lower boundary of the detectable range."

RC1.23: P17, L452: "dust particles"

AC1.23: Following the suggestion, the text has been updated (L452, L561 tracked changes).

"In a recent study, Cornwell et al. (2020) have shown that re-emission of dust particles from sea water into the atmosphere is possible and that the re-emitted particles retained their ability to act as INP."

RC1.24: P17, L457: "can not" -> "cannot" (there are other instances)

AC1.24: This sentence has been removed (L455, L565 tracked changes). However, another part of the text has been updated accordingly (L502, L634 tracked changes).

"Note that contrary to sodium and MSA (see subsection 2.2), an influence of the RV's ship exhaust on PM_{10} mass cannot be ruled out."

RC1.25: P17, L462: "presented", see above.

AC1.25: Please see AC1.9

RC1.26: P17, L464: "Fig.7c-e"

AC1.26: We changed the reference accordingly (L464, L573 tracked changes).

"The respective PDF of $N_{INP,-16}$, $N_{INP,-12}$, and $N_{INP,-8}$ are included for comparison in Fig. 8c-e."

RC1.27: P18, L492-494: As noted above, this argument could already be mentioned earlier in the manuscript.

AC1.27: Please see AC1.3

RC1.28: P19, L536: Maybe: "Another factor causing year-to-year variability could be the presence of sea ice."

AC1.28: The text has been changed following the suggestion (L536, L670 tracked changes).

"Another factor causing year-to-year variability could be the presence of sea ice."

RC1.29: P19, L537: Delete comma after "ACE".

AC1.29: The text has been updated accordingly (L536, L670 tracked changes).

"Schmale et al. (2019) note a significantly lower sea ice extent on the Amundsen Sea during ACE when compared to climatological records."

RC1.30: P19, L538-540: This sentence is also nested and reads a bit complicated. Please reformulate.

AC1.30: The sentence was reformulated to improve readability (L538, L674 tracked changes).

"Additionally, variations in atmospheric MSA sink strength are a potential contributor to variability in observed MSA mass concentrations. For example, MSA is efficiently removed from the atmosphere by precipitation. In the SO, rain events are associated with frontal zones."

RC1.31: P20, L553: Delete comma after "both".

AC1.31: The text has been updated accordingly (L552, L688 tracked changes).

"As sodium is used as a conservative tracer for primary aerosol particles of marine origin (Legrand and Pasteur, 1998), especially sea salt, the correlations suggest that SSA significantly contributes to both PM_{10} and the coarse mode."

RC1.32: P20, L556: Delete comma after "mentioning"

AC1.32: This part has been completely revised (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)."

RC1.33: P20, L557: Delete both commas in the sentence starting with "Another factor"

AC1.33: The text has been changed accordingly (L557, L694 tracked changes).

"Another factor might be that the wind speed was averaged over 24 h in order to match the temporal resolution of the filter sampling."

RC1.34: P20, L559: Delete comma after "Note"

AC1.34: This part has been completely revised (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 $\pm 70\%$."

RC1.35: P20, L562: "atmospherically" relevant

AC1.35: The text has been updated accordingly (L561, L699 tracked changes).

"The particle concentration of mode 2 shows a positive correlation ($\rho \sim 0.7$) with N_{CCN} at $SS \leq 0.5$ %, pointing at the importance of accumulation mode particles for the CCN population at atmospherically relevant SS."

RC1.36: P20, L571-573: Again a rather long sentence that could be split into two parts.

AC1.36: The sentence was split to improve readability (L571, L711 tracked changes).

"Considering the long process chain from in-water DMS to particles of CCN size, this is not surprising. The argument in Bates et al. (1998), that conditions at measurement point may not be representative for the conditions encountered by the air-parcel during transport, is applicable here as well."

RC1.37: P21, L594: Maybe better "total number-based or total mass-based"

AC1.37: The sentence was changed accordingly (L594, L742 tracked changes).

"This clearly shows that deriving INP-related properties from total number-based or total mass-based aerosol properties without considering air-mass history might lead to results not representative for atmospheric N_{INP} ."

RC1.38: P22, L625/626: See above, the findings from Schmale et al. (2019) (aren't they also based on a subset of the ACE CCN data?) could already be shortly addressed in the introduction.

AC1.38: Please see AC1.2

RC1.39: P22, L647: "a unique"

AC1.39: The sentence was changed accordingly (L647, L801 tracked changes).

"The presented data set gives a unique, circum-Antarctic view on CCN and INP abundance, their properties and indications towards aerosol particle origin."

RC1.40: P22, L651: "through"

AC1.40: The text has been updated accordingly (L651, L805 tracked changes).

"The ACE data are available through web portal ZENODO, with INP data from LV sampling under <https://zenodo.org/record/4311665> (version 1.1), CCN data under <https://zenodo.org/record/4415495> (version 1.1), and data on MSA and sodium under <https://zenodo.org/record/3922147> (version 1.0)."

RC1.41: P33, last but one line: "are" indicated

AC1.41: The caption of Fig. 8 has been updated, accordingly (P33, P23 tracked changes).

"The number of data points (n) are indicated in the figure."

RC1.42: Caption of Fig. S4: Wrong order of the panels; duplicate "the" in the last but one line.

AC1.42: The caption of Fig. S4 has been updated accordingly (see below).

"INP number concentration at (a) -24, (b) -20, (c) -16, (d) -12, and (e) -8°C for the LV filters sampled during ACE (circles)."

RC1.43: Caption of Fig. S5: Replace "and corresponding factor two" by "± a factor of two" as in Figure 8.

AC1.43: The caption of Fig. S5 has been updated as suggested (see below).

"Average spectra of field blank filters (FBF) ± a factor of two (pink line and area) and data range from McCluskey et al. (2018) (light blue envelope) are given for reference."

RC1.44: Table S2: What do you mean by "during parts" of the cruise? No Leg-resolved data are shown. You could also better describe the two different approaches for averaging as in lines 422-424 of the article.

AC1.44: Thank you for your comment. The caption of Tab. S2 has been updated to clarify the differences in averaging (see below).

"Overview of the LV sampling INP number concentrations ($N_{INP,LV}$) measured throughout the cruise, given as mean, median, and geometric mean values (including one geometric standard deviation)."

RC1.45: Table S4: See above, what do mean by "during parts"?

AC1.45: The caption for Tab. S4 has been updated to clarify (see below).

"Overview of the HV sampling INP number concentrations ($N_{INP,HV}$) encountered throughout the cruise, given as mean, median, and geometric mean values (including one geometric standard deviation). Averaging was performed with the inclusion of concentrations on the lower/upper boundaries of sensitivity and the number of samples are indicated (n) in the table. Differences in sampling strategy for HV compared to LV samples can be found in subsection 2.3."

RC1.46: Table S5: You could chose "M" as symbol for the mass concentration.

AC1.46: This is a good idea. The mass concentration values have been denoted throughout the text by M .

Please also note the supplement to this comment:

<https://acp.copernicus.org/preprints/acp-2021-700/acp-2021-700-AC1-supplement.pdf>