Response to Reviewers

Long-term Variability in immersion-mode Marine Ice Nucleating Particles from Climate Model Simulations and Observations

Aishwarya Raman et al.

RC1: 'Review of "Long-term Variability in immersion-mode Marine Ice Nucleating Particles from Climate Model Simulations and Observations" by Raman et al.', Anonymous Referee #1, 10 May 2022

General comment:

Reply: We appreciate the referee's valuable comments.

Reviewer comment: Simulated concentrations of ice nucleating particles (INP) are compared to long-term field observations from the Southern Ocean collected during the MICRE campaign. INP concentrations are calculated using various parametrizations that link simulated sea spray and dust aerosol concentration or surface area to the abundance of INP. From the comparison with ambient data, an underprediction of aerosol by the E3SM model is identified. It is concluded that due to the lack of parallel observations of aerosol properties during MICRE, which would allow verification of the simulated input parameters for the INP parameterizations, the comparison with the INP observations is inconclusive and the reasons for model-observation differences cannot be quantified using the MICRE dataset. Many of the manuscript's insights appear in previous work and are echoing the recent review by Burrows et al., 2022.

Reply: I briefly describe our motivation for using the MICRE dataset.
Previous studies have found large climate model biases in high latitudes and these studies suggested improvements to the model representation of INPs can reduce the model bias in cloud radiative properties and cloud phase (Vergara-Temprado et al., 2018, Tan et al., 2019, Tan et al., 2022). Despite the importance of INPs, so far, over remote marine regions, INP modeling studies have analyzed only short-term INP datasets and compared them to observed INP concentrations. These studies were limited by the temporal coverage of measurements (1 week – 1 month) (e.g. McCluskey et al., 2019). While these studies provided important insights into the model-observation comparisons of INPs and sources of INPs during this time, the applicability of INP parameterizations is sensitive to the time period of the campaign, instruments used for INP characterization, and geographic location. For example, the primary source of INPs in continental regions is different from the background INPs in remote marine regions.

The objective of this study is to explore the utility of a long-term INP concentration dataset from the MICRE campaign to evaluate climate model simulations of INP concentrations in the Southern Ocean (SO). This is a more direct comparison between simulated and observed studies for a full year's worth of INP observations and is a step forward from previous studies which have either looked at short-term INP datasets or indirectly analyzed INPs by probing the model sensitivity in the cloud or radiative properties due to changes in INP concentrations.

We agree that the lack of aerosol measurements is one of the limitations of this study because that limits our ability to understand and associate the bias in INPs with bias in simulated aerosols. But, this does not invalidate the knowledge gained using the MICRE dataset and the climate model simulations on model-measurements gaps for INPs over the SO. The findings from this study can

be useful for various future studies assessing model biases in the SO climate and also help design future INP and aerosol campaigns in this region because this study also highlights the measurement needs.

Although Burrows et al., 2022 is a relevant paper that provides an overview of observational and modeling needs for INPs, the current study, 'Long-term Variability in immersion-mode Marine Ice Nucleating Particles from Climate Model Simulations and Observations', is a specific model-observation comparison study in the SO using the MICRE campaign INP dataset.

Reviewer comment: The MICRE dataset is an inadequate choice for the intended method of comparison. Therefore, the exercise should be repeated using a different field data set (for example the Tatzelt et al. 2022 data from the SO that includes aerosol measurements). An alternative approach could be to focus on the variability of INP in the SO and the usefulness of the aerosol-aware parametrizations to reproduce such. For example, the MICRE dataset is very valuable as it demonstrates the absence of a strong annual variation. The lack of seasonal variation could indicate that SO INP sources have little seasonal dependence and are likely not sensitive to global warming. Concentrations are more variable on shorter time scales. It could be examined whether the input parameters for aerosol-aware parametrizations reflect this variability on the time scale of days, months, or seasons. It should be elucidated how much of the observed variability in the MICRE observations, detected in sample volumes of 10s of m^3 on time scales of days, is expected to be mirrored in the simulation with billions of m^3 per grid cell and a time step of 30 min. Focusing on metrics of variability in time and space and comparing them between the simulation and observations could make better use of the unique MICRE dataset.

Reply: We appreciate the reviewer's suggestions for additional INP datasets in the SO. Following the reviewer's comments, we have made several additions to the manuscript. We would also like to clarify the model-observation colocation methods that were performed to compare the observed and simulated variability in INPs.

We added statements about the co-location in the manuscript in lines 237-239. We co-located the model with the MICRE INP dataset spatially (nearest grid cell in the model to Macquarie Island location) and temporally (taking the average of the same days used for INP calculations from observations) which is common in most model-measurement comparison studies.

Following up on the reviewer's suggestion to include other INP datasets in the SO, we have added a comparison of the MICRE dataset to two other ship-based campaigns in the SO, MARCUS, and ACE. We have included these results in Figure 8 and Figure 10. Figure 8 shows the probability density functions of INP measurements from MICRE, MARCUS, and ACE (closer to Macquarie Island) and simulated INPs from different aerosol-aware INP parameterizations.

We have added the number of data points used to construct the PDFs in the last sentence of the caption for Figure 8. In Panel (a) of Figure 10, we show MICRE INP-temperature dependency along with other observations from MARCUS (ship measurements in the open ocean and closer to Macquarie Island) and ACE (cruise leg that was closer to Macquarie Island. These figures provide a comparison between MICRE INPs and INPs from other campaigns in the SO.

Additionally, the scatterplots in Figure 5 provide metrics such as modified normalized mean bias, correlation coefficient (variability metric), and fractional gross error which indicate how modeled INP concentrations compare to observations for different temperatures for the campaign period.

Figure 6 also shows time series plots that help understand how model variability compares to the observed variability in the time scale of days.

In addition to these, we have also added three separate sections 3.6.1 – 3.6.3 to discuss the bias in modeled INP concentrations and the interpretation of model bias. We also performed idealized simulations of frozen fractions for different INP conditions and for different measurement approaches (isothermal and constant cooling-rate experiments) to demonstrate how the time-dependency of frozen fractions behaves for different measurement approaches at different temperatures. This provides additional information on how to interpret observed and modeled INP concentrations based on different instrument configurations.

Specific comments:

Line 38 ff.: Droplets do not freeze heterogeneously at lower supersaturations if they contain INPs.

Reply: Line 43

We removed the phrase supersaturation because it is relevant to deposition nucleation and not immersion freezing. "In the presence of aerosol particles that can nucleate ice such as dust, bacteria, and fungal spores, supercooled cloud droplets can freeze into ice at warmer temperatures than required for homogeneous ice nucleation (Vali et al., 40 2015; Vergara-Temprado et al., 2018; Kanji et al., 2017)."

Line 65, 68, 72: Define what is meant by INP efficiency. For example, onset temperature, active site density, ice nucleation rate.

Reply: Line 69
We have clarified INP efficiency. We refer to INP efficiency using ice active-site density. "Laboratory experiments using realistic proxies for biologically-influenced sea spray particles have clearly shown that these particles can act as INPs, although IN efficiency of dust (expressed in terms of ice active-site density ns) is ca. 2 orders of magnitudes more than sea spray particles for a given aerosol surface area and for temperatures colder than 263 K (DeMott et al., 2016; McCluskey et al., 2018b)."

Line 75ff: Is a lot of NH dust e.g., from Nord African transported to the SO? Dry areas in the SH, particularly Southern Africa seem more plausible sources.

Reply: Line 80

We corrected the typo and changed it to South Africa. "The source regions of dust transported to the SO include South America, Australia, New Zealand, and South Africa (Wagener et al., 2008; Struve et al., 2020; Neff and Bertler, 2015). "

Line 93: clarify how the "representativeness of field measurements" can be evaluated. Any observation is representative of the time and place it is conducted.

Reply: Lines 96-98
We have rephrased the sentence as follows: "Without such observations, it is challenging to understand and constrain the aerosol sources and processes driving seasonal INP variations in climate models"

Line 101: quantify "temporally representative"

Reply: We rephrased lines 106-110 as follows.
"Few studies have systematically (co-located in space and time) evaluated the simulated INP concentrations from climate models in the SO. Such a comparison using long-term INP observations is critical for assessing the seasonality of different INP sources and their impacts on SO clouds and energy budget."

Line 174, Tab.S1: Add what aerosol property (surface area, mass, number) the parametrizations use. The formulas of the parametrizations could also be included.

Reply: We have added an additional column in Table S1 in the Supplementary Text that displays the aerosol property used in different INP parameterizations along with the units.

Line 184: check unit of Jimm,dust. Should it be [s^{-1}]? Is the CNT scheme independent on particle size?

Reply:
Lines 186-195

We have corrected the typo in the units for Jimm, dust and also included the particle size dependency for Jimm_dust in Equation 1 and 2.
In this study, we use INPs from E3SM CNT calculated using the heterogeneous freezing rate (Jimm_dust) as a function of ambient temperature and model's modal radii of dust in accumulation and coarse mode (80 nm – 10 μm).

Lines 190 – 195 – Particle size dependence

– "where Naer,dust [m^{-3}] is the total (cloud-borne and interstitial) dust number concentration in accumulation and coarse modes (ice-borne aerosol is not tracked in the model), and Jimm,dust [s−1] is the heterogeneous nucleation rate for dust calculated as a function of modal radii of dust, r_{aer}, and model's ambient temperature,$T_{ambient}$. Analogous relationships are implemented for each ice-nucleating species and freezing mode handled by the CNT scheme."

Line 185 f.: Are other aerosol species than dust included in the CNT scheme? Please list their nucleation rates and give references.

Reply: The E3SM model also includes black carbon in the CNT scheme. However, recent studies indicate that black carbon contributes negligibly to immersion freezing in the atmosphere. So, we exclude that in the current study and include only dust aerosols for calculating INPs from the CNT approach.

Line 197: 10s is typical for CFDC instruments. The cooling rate in CSU-IS could be used to estimate the timestep for this type of experiment which is probably closer to 60s.

Reply: We acknowledge the importance of time dependency in the CNT approach. To compare the E3SMv1 model's CNT-based parameterization with INPs observed by the ice spectrometer, it is necessary to make assumptions, either implicit or explicit, about the time dependence of immersion freezing.

To address the reviewer's comment, we performed a series of idealized immersion freezing experiments to compare E3SM INP 10s diagnostic for different measurement configurations. We calculated INP for isothermal experiments with different residence times and constant cooling-rate-dependent experiments with different cooling rates. We repeated these experiments for different ice nuclei surface area distributions (or INP abundance).

We discuss these results in Section 3.6.1 'Bias in E3SM CNT INP concentrations. The list of experiments is given in Table 5.

For low INP conditions like those typically observed in Macquarie Island, we find that the E3SM linearized diagnostic for 10s is in good agreement with the idealized frozen fraction experiment corresponding to the MICRE cooling-rate of 0.33 K/min. For this case of low INP conditions, E3SM diagnostic is also in good agreement with the isothermal experiments with 10s residence time. However, this diagnostic underestimates INP concentrations in high dust conditions in comparison with isothermal and constant cooling-rate experiments.

We recommend that the time step dependence of INP should be further explored in future studies of this kind. The treatment of INPs in E3SM and model development is beyond the scope of this study.

Line 210: The ice spectrometer measurement starts at 0°C and not -5.1°C. The latter is probably the highest temperature where freezing was detected.

Reply: Line 218
We have revised the text to read "Filters were processed using the Colorado State University ice spectrometer (McCluskey et al., 2018a) to obtain temperature spectra of immersion freezing INP concentrations from - 28◦C to 0◦C (DeMott et al., 2018)."

Line 218 ff: In DeMott et al. 2018 the potential use of sun photometer and BOM lidar data to retrieve aerosol surface area during MICRE is mentioned. Could this help the analysis? As mentioned in the general comment, a large SO dataset from the ACE campaign (20.12.2016-19.3.2017) taking place right before MICRE is available that also contains several aerosol measurements.

Reply: We have included the ACE dataset in Figure 8 and Figure 10. We have also added relevant discussions for comparing MICRE and ACE in Figures 8 and 10. Tatzelt et al., 2021 showed that there is no significant correlation between INP concentrations and measured aerosol mass concentrations.

There is a separate study by Paul J. Demott's group that is looking at the sun photometer and BOM lidar data during MICRE which is beyond the scope of this study.

We are not aware of co-located INP and aerosol size-resolved composition measurements in the SO that coincide with the MICRE campaign. If the reviewer is aware of other aerosol size distribution datasets and co-located INP measurements in the SO, we would be happy to consider that for further analysis.

Line 235: How are freezing rates simulated? They seem not to be used in this study.

Reply: We calculate the heterogeneous nucleation rate for dust INPs in E3SM following Equation 1 in Hoose et al., 2010. In E3SM, the freezing rate is calculated by dividing Equation 2 by the model timestep. In our case, we multiply the E3SM calculated freezing rate by the measurement or assumed timestep to derive INP concentrations. Freezing rates calculated in E3SM are not directly used in this study because the goal is to compare simulated INP concentrations against the measurements.

Line 248: The time period does not agree with the dates in Tab.1.

Reply: We thank the reviewer for pointing us to this typo. We have corrected it.

Line 246 ff: Additional sources of long-term measurements in the SO could be data from the Australian "Atmospheric Baseline" program and measurements taken on board the annual supply ships to the Antarctic stations.

Reply: Presently, no long-term aerosol measurements are collected aboard the Australian icebreaker (RSV Nuyina), which regularly transits the SO during summer to resupply the Australian bases. However, plans are underway to provide this capability in the future.

Section 3.1. The particle number size distribution is more relevant for the tested INP parametrizations than aerosol mass concentration (except W15). A test of the prescribed size distribution against observations could be more conclusive to investigate the simulated aerosol input parameters.

> **Reply:** It would be interesting to see if there are any long-term measurements of aerosol size distribution with co-located INP measurements in the SO. We are aware of short-term campaigns. We cannot use them for closure comparisons. We are not aware of long-term measurements of aerosol particle number size distribution that cover coarse mode dust and sea spray and also match the time period when INP measurements were collected.

Sections 3.1, 3.2 The model analysis is not specific to this paper. It seems after Sec. 3.1 the requirements for a comparison dataset become obvious but are not considered for the choice in Sec. 3.3. Sec. 3.2 could be removed without effect on the outcome of the study.

> **Reply:** Section 3.2 discusses the simulated global mean INP distributions from different aerosol-aware INP parameterizations. This section provides insights into how INPs in the SO compare against other regions and how different INP parameterizations compare against each other in terms of different geographic regions and at different temperatures.
>
> We find that in general INPs in the SO are several orders of magnitude lower than the continental regions or mid-latitude oceans. So, INP parameterizations developed for continental regions cannot be directly applied to predict marine INPs in remote regions like the SO. Therefore, the applicability and limitations of these parameterizations have to be carefully considered in model observation comparisons and for the global modeling of INPs.

Line 313: Elaborate the implications of the model evaluation.

> **Reply**: Most of the INP measurements are near the surface. Evaluating the vertical profiles of aerosols from ATom measurements helps to assess if simulated bias in INP concentrations are mainly due to the biases in the model transport and removal mechanisms of aerosol particles. Burrows et al., 2022 also discusses the need for vertically resolved INP concentrations and how significant biases in INP concentrations are linked to aerosol transport in the model.
>
> Following the reviewer's comments, we have added the following implications of the model evaluation for vertical profiles of aerosols.
>
> Lines 318-322: "While the MICRE observations were conducted in near-surface air, the impacts of INPs on clouds occur at higher altitudes. Therefore, adequate simulation of dust and sea spray aerosols above the surface level is also required to correctly simulate the impacts of INPs on cloud properties. Evaluating the vertical profiles of aerosols from model and observations helps to identify if simulated biases in INP concentrations are mainly due to the biases in the model transport and removal mechanisms of aerosol particles or due to the aerosol emissions near the surface (Burrows et al., 2022)."
>
> Lines 344 – 347: "The model evaluation of vertical aerosol profiles has implications for the role of INPs in cloud optical and microphysical properties in the MBL and free troposphere (Murray

et al., 2021; Tan and Storelvmo, 2019; Burrows et al., 2022). Using lidar retrievals of dust, smoke, and cloud phase, Tan et al. (2014) showed that the presence of dust and smoke particles were negatively correlated with the supercooled liquid fraction."

Figure 4: Contrary to the comparison to data from measurement stations at surface level the model seems to overpredict the dust and sea salt mass by a factor >2 compared to the PALMS data. Ship-based datasets could provide higher resolved information on spatial variations.

Reply: While ATom observations are for a few days in different years, we note that model values are interpolated along the flight track. In the revised version, in Figure 4, for each pressure level, we also added standard deviation for model dust and sea salt concentrations which show that E3SMv1 dust is mostly within the standard deviation of PALMS measurements.

Lines 336-339: We elaborate on why cannot directly compare the model performance for E3SM – in situ measurements against PALMS-model agreements.

" The interpretation of model-observation agreement in vertical profiles should be taken with caution because the Atom measurements used in Figure 4 includes only eight days of flights (dates are provided in Figure 4 caption) and might only be representative of zonal average aerosol concentrations for the flight days. However, E3SM-simulated dust and sea salt concentrations represent monthly averaged values for the flight track. This is likely one of the reasons why simulated dust concentrations in E3SMv1 show an underestimation of dust compared to in situ dust climatologies at SH stations (Figure 2), but are within the observational uncertainty for ATom measurements. Although the ATom flights did not directly pass over 340 Macquarie Island, these comparisons are useful to understand the model's general behavior in simulating the vertical profiles of dust and sea salt concentrations in this region. Good agreement in model vertical gradient in dust and sea salt aerosols indicates that vertical mixing is likely a smaller concern. The agreement between model and observations was assessed by checking if ATom observed mean values are within the simulated standard deviation for dust and sea salt."

Line 337: Clarify what the implications are.

Reply: }. We thank the reviewer for the suggestion. We have rephrased the sentences as below.

Lines 373 - 382

"This assumption may have important limitations; e.g., differences in the size distribution and composition of INEs released into atmospheric SSA particles due to the different SSA production mechanisms (Wang et al., 2017). A recent study using laboratory measurements showed that the INP concentrations from submicron-sized SSA were lower by a factor of 10 compared to atmospheric INP concentrations from total SSA (Mitts et al., 2021). This study found that in addition to the submicron INEs within the SSML, supermicron-sized SSA particles produced from jet drops were also important to the total marine INP concentrations. Using the cruise measurements, Trueblood et al. (2021) investigated the relationships between SSML INP and SSA INP at $-15^{\circ}C$ during a dust wet deposition event in the Mediterranean Sea. These observations showed a three-day lag between the increase in INP concentrations in the SML and the increase in INPs from total SSA. This study concluded that processes governing the evolution of INPs in SSA and INPs in SSML are not the same. The effects of

INEs and INPs on clouds in remote marine regions will therefore vary based on their composition and other physical properties. "

Table 3 and 4: Comparing to globally averaged field data would be informative, e.g., Kanji 2017 Fig.10 and Welti 2020 Fig.5 for D15, CNT and M18, W15, respectively.

Reply: Lines 390 - 395

Following the reviewer's comments, we added a paragraph comparing the global mean values from E3SM in our study against Kanji et al., and Welti et al., in the subsection, Simulated global mean INP distributions.

"Kanji et al. (2017) compared temperature-INP spectra from studies of field measurements conducted globally for different categories of INP composition. This study found several orders of magnitude differences in INP concentration within any air mass or particle composition. Figure 10 in Kanji et al. (2017) shows that at $-20^{\circ}C$, INP concentrations for marine samples range from 0.001 L^{-1} to 2.0 L^{-1}. The SH INP number concentration mean from D15, The E3SM-simulated SH INP mean across the temperature spectra are also within the range of most frequently observed INP concentrations (\SIrange{0.0001}{0.1}{\per \liter}) shown in Figure 5 of Welti et al. (2020) for the South Polar marine regions."

Line 362: In comparison to what is the agreement of W15 better?

Reply: We rephrased this sentence as follows in line 413

"Interestingly, the W15 parameterization also produces better agreement with observed INPs compared to M18+D15."

Line 366: DeMott 2018 p.3 mention that chemical and biological analysis have received funding. Will this data become available in the future?

Reply: Demott et al., and his group are working on a separate paper for data analysis of MICRE. Commenting on a future chemical and biological analysis dataset is beyond the scope of this study.

Figure 6: Add FGE to the caption. Is NMB equivalent to MNMB introduced on line 262? Discuss what are the different skill scores reveal.

Reply: Lines 280-290
We added FGE to the caption in Figure 6. We clarified the modified normalized mean bias (MNMB) and Fractional Gross Error (FGE), added a new paragraph describing the evaluation metrics, and added Equations for FGE and MNMB (Equations 3 and 4).

"To assess the predictive skill of simulated INP concentrations in near-surface air, we use a set of standard skill scores outlined by the Monitoring Atmospheric Composition and Climate (MACC-II) project model evaluation methods (Cuevas et al., 2015; Eskes et al., 2015; Huijnen and Eskes, 2012). The modified normalized mean bias (MNMB), Pearson's correlation coefficient, fractional gross error (FGE), and percentage of data points within a factor of 2 (2x) and 10 (10x) from the observations are used to assess E3SMv1 INP concentrations. McCluskey et al. (2019) used MNMB and FGE to assess the model's ability to simulate INP concentrations observed in Mace Head and the SO. We define MNMB and FGE in Equation 3 and Equation 4 respectively. In both these metrics, the differences between the observed and predicted INPs are normalized by the sum of observed and predicted INP concentrations. Both are symmetric; MNMB ranges from -2 to 2 and FGE ranges from 0 to 2. Because they are normalized, both these metrics avoid enormously high values in cases where model-observation differences go up to several orders of magnitude."

Figure 7: Explain the numbers given in the figure legend in the caption.

Reply: Following the reviewer's comments, we add the following statements in the caption. "The values displayed in the brackets in the legend show the mean values from observed and simulated INP concentrations"

Figure 7 caption: In what way are simulated INP interpolated? Over time? An indication of variation for the simulated INP would be helpful.

Reply: Following the reviewer's comments, we add the following statements in the Figure 7 caption. "Simulated INP concentrations are interpolated to the grid box closest to Macquarie Island using the nearest neighbor interpolation method. For each sampling period, simulated values are output as instantaneous values every 30 minutes and averaged from the start date to the end date. "

Section 3.4.: Other long-term INP field studies (e.g., Schrod et al., 2020; Welti et al., 2018) have shown that INP concentrations correlate poorly or not at all with bulk aerosol measurements due to the rarity of INP. They also showed weak annual trends. Contrary to what is suggested in the manuscript, it could be concluded that due to low seasonal variability, local INP concentrations can be measured representatively during a multi-week campaign and that the most important task to achieve causal aerosol-aware INP parameterization is to identify INPs that are active at different temperatures at the particle level (also suggested in Burrows et al., 2022) rather than measuring bulk.

Reply: There is a separate study underway on the analysis of seasonal variability in MICRE INP concentrations and correlating them to possible seasonal INP sources and mechanisms by Hill.T et al. We agree that measuring size-resolved aerosol composition is the most important task to understand and casually associate the variability in INPs to processes and INP sources and our respective research groups are working on this in other separate efforts. A multi-week campaign that measures INP and aerosol size distribution will be helpful to identify INPs active at different temperatures.

The annual trends in INP concentrations are different based on the region, geography, and seasonal sources. Welti et al., 2018 analyzed relations between total particle mass concentration and IN. This study suggested that the INP concentrations can show a higher statistical correlation with bulk aerosol properties such as the total particle mass concentration only during episodic aerosol events such as forest fires (McCluskey et al., 2014) and dust storms (Boose et al., 2016).

Therefore, investigating the relationships between aerosol mass or number concentration and total INP concentrations cannot be always the best strategy to identify the sources of INPs. We would also like to note that none of the sites analyzed in Schrod et al., 2020 were in the SO. This study also primarily analyzed the relations between bulk aerosol properties such as aerosol number concentration and INPs. Burrows et al., 2022 pointed out that size-resolved aerosol composition is necessary to understand the contribution of different aerosol sources to INPs at different temperatures.

Long-term measurements such as MICRE show seasonal variability in INPs and therefore provide an opportunity to understand the sources in high and low INP days. Measuring INPs in different seasons also helps to capture INP sources during seasonal episodic events such as dust storms or forest fires. However, there could be regional differences in the contribution from specific sources, INP composition, annual INP trends, and the relations with bulk aerosol properties based on transport patterns, aerosol characteristics, geographical climate, and ecosystem.

We have included the reviewer's suggestions in the following paragraph in lines 619-628.

"On the other hand, other long-term INP field studies (Schrod et al., 2020; Tatzelt et al., 2021; Welti et al., 2018) have shown that bulk aerosol concentrations do not necessarily correlate with INP climatology. A recent work by Creamean et al. (2022a) also showed that there was no significant relationship between the INP concentrations and particle size in the central Arctic for year-long size-resolved INP observations. However, due to the lack of in situ aerosol measurements during MICRE, we could not conclusively attribute the causes of the model-observation differences or explain the sources of day-to-day variability in measured INPs, although a closer investigation of this issue is the topic of a separate, ongoing study. While a multi-week campaign with simultaneous measurements of INP, particle chemistry, and particle size distributions can improve the predictability of INP concentrations, long-term INP measurements such as MICRE are important to understand the impacts of synoptic weather and seasonal aerosol characteristics on INP concentrations."

Section 3.5.: The PDF from the field measurements refer to much smaller air volume than the model. Please explore possible biases and show that such a comparison is valid.

Reply: The model values were averaged for a grid box (1^{o}x1^{o}) nearest to Macquarie Island. Such interpolation of model results to compare against point-based measurements in common in the climate and air-quality communities. To address the reviewer's comment about the air volume sampled in the model and measurements, we have added the number of measurements used at each temperature. Model INPs were averaged from the start date to the end date for each sample during the measurement period.

Figure 8: The PDFs go to extremely low concentrations not seen in Figs. 6, 7 and 9. Please double check if the values are correct and specify what data (location, timestep) is used for the plot.

Reply: We have revised Figure 8 following the reviewer's comments. We also added the probability density functions for additional INP datasets in the SO to compare MICRE INP distributions against other SO INP datasets.

Technical corrections:

Some citations seem to satisfy key words instead of content. If the citation is specific to a finding in a paper it would be helpful to give a one-line summary of the finding, otherwise an effort should be made to track original or most comprehensive sources of concepts.

Reply: We thank the reviewer's suggestions for references. We have revised the citations throughout the paper following the reviewer's comments.

Line 41: Vali 2015 would be a better reference for INP.

Reply: Line 45:
We have corrected this reference. "The aerosol particles responsible for this process of heterogeneous ice formation are called ice nucleating particles (INPs) (Vali et al., 2015)."

Table S1, line 519: should it be mineralogy?

Reply: We have changed the text in Table S1. "Potential variations in marine INPs due to ocean biology or dust mineralogy are not captured."

Line 116-117: correct citation style

Reply: We have corrected the citation style.

Line 213: Vali 1971 would be a better reference.

Reply: Line 222
We have added the reference suggested by the reviewer. "Concentrations of INPs were calculated at different temperatures using the fraction 220 of unfrozen wells per given temperature (Beall et al., 2017; Vali, 1971)."
"

Line 284: …whereas it overestimates…

Reply: lines 301 – 303
We revised the typo in these lines. "Turning to sea salt, Figure 3 shows a model overestimation by at least an order of magnitude at Mawson and McMurdo Station in Antarctica whereas it underestimates sea salt in Cape Grim and Palmer Station. However, the model underestimates sea salt climatology by 1–2 orders of magnitudes in the NH stations (Figure S2)."

Line 285: Missing space after comma. Error in the figure number.

Reply: Thanks, we have fixed the missing space and the figure number.

Line 289: correct citation style

Reply: Line 310
We have corrected the citation style in the revised version "deposition is a major loss process for supermicron aerosol, and the parameterization of dry deposition used in E3SM was recently shown to overestimate deposition to the ocean (Emerson et al., 2020)."

Line 295: missing closing bracket

Reply: We have added the closing bracket in the revised version in lines 313 and 319.

Figure 2, 3: homogenize axes in (e) with the other subfigures.

Reply: Following the reviewer's suggestion, we have revised the axes in Figures 2 and 3 and in the supplementary text, figures S1 and S2.

Figure S4 (a), (b): Homogenize axes and subfigure size. Include explanation of colours in (a) to caption.

Reply: We have homogenized axes and subfigure size in Figure S4. We also explained the colors in the caption for Figure S4.

Table 3: M18 instead of M17

Reply: We have changed M17 to M18 in Table 3 in the revised version.

Line 387: correct citation style

Reply: We have corrected the citation style in the revised version in line 434.

Figure 8: Homogenize font size. This should be done more carefully in all figures.

Reply: We have revised Figure 8 and corrected the font size.

Line 415: remove line break

Reply: We have rephrased several paragraphs in this section and added/removed the necessary line breaks.

Line 420: Jumping topic. Add line break.

Reply: We have rephrased several paragraphs in this section and added/removed necessary line breaks.

Line 441, 500, 511, 513: correct citation style

Reply: We have corrected the citation style in the revised version in lines 501,630. We have rephrased citations in Lines 511 and 513 in section, 3.6.2 'Bias in E3SM simulated aerosol properties'.

Code availability: remove "also"

Reply: We have corrected this in the revised manuscript version.

Reviewer 2

We appreciate the reviewer's valuable comments. We have revised the manuscript accordingly and replied to the reviewer's specific comments below.

Line 264: Can a reference please be provided for ATom PALMS instrumentation and measurement?

Reply: In the revised version, we added the following reference in the revised version in line 272.

Froyd, K. D., Murphy, D. M., Brock, C. A., Campuzano-Jost, P., Dibb, J. E., Jimenez, J.-L., Kupc, A., Middlebrook, A. M., Schill, G. P., Thornhill, K. L., Williamson, C. J., Wilson, J. C., and Ziemba, L. D.: A new method to quantify mineral dust and other aerosol species from aircraft platforms using single particle mass spectrometry, Atmos. Meas. Tech., 12, 6209–6239, https://doi.org/10.5194/amt-12-6209-2019, 2019

Figure 2: Please format the ordinate axis in panel(e) to that of the other panels.

Reply: We have formatted panel (e) in Figures 2 and 3 and homogenized the axes.

Figure 2 caption: Can you please clarify "Also shown are the E3SM simulated sea salt climatology at the Macquarie Island and time series of dust concentrations from AWARE field campaign with co-located E3SM model simulations"? I am only seeing the observed and simulated dust climatologies in this figure, not sea salt.

> **Reply:** We have corrected the typo in the Figure 2 caption. The revised version reads, "Climatology of dust concentrations from ground stations in the Southern Hemisphere (SH) compared against E3SMv1 aerosol climatology. Ground stations and their locations are listed in Table 1. Model aerosol concentrations were derived from monthly average dust concentrations for the period 2016-2018. Error bars in the model represent standard deviation of dust aerosol mass concentrations for 2016-2018. Error bars in the observations for each ground station represent standard deviation of measurements for the period shown in Table 1. Both CTL and sensitivity simulations (EXP) are shown for comparison with observations. Also shown are the E3SM simulated dust climatology at Macquarie Island and time series of dust concentrations from the AWARE field campaign with co-located E3SM model simulations."

Line 285: Which figure is being referenced here? Is it Figure S2? (authors have left "??").

> **Reply**: We have corrected the figure number in the text in the revised version.
> "However, the model underestimates sea salt climatology by 1–2 orders of magnitudes in the NH stations (Figure S2)"

Line 285: This line reads a bit awkwardly; please proof read. A "the" might need to be added following the comma after "stations".

> **Reply: Lines 305-306** We replaced this sentence with "However, the model also underestimates sea salt climatology by 1--2 orders of magnitudes in the NH stations (Figure S2)" in the revised version.

Line 290: change "budget" to "budgets"

> **Reply**: Line 311: In the revised version, we changed "budget" to "budgets".

Line 303-305: Please clarify, "dust concentrations do not vary much below 400 hPa in E3SM simulations and ATom measurements". Do the authors mean the difference in values between the model and observations are not very different? Or, are the authors discussing a statistical metric of variability in the average values below 400 hPa; no model variability is shown in Figure 4.

> **Reply:** We refer to the standard deviation in model and measurements in these sentences while discussing the differences between Atom and E3SM vertical profiles of dust.
> Lines 327 – 335.
>
> Following the reviewer's suggestion, we have rephrased the paragraph as follows:

"Figure 4 shows that E3SMv1 adequately simulates the dust and sea salt concentrations to within the range of observed values at all pressure levels up to 400 hPa, with the exception of 800 hPa where the number of observations are too small to provide a meaningful sample (N = 2). ATom measurements are converted to concentrations 330 under standard temperature and pressure. We find that the simulated dust concentrations from E3SM (Figure 4, red lines) shows smaller standard deviation compared to that observed in ATom measurements (Figure 4, grey lines). In contrast, sea spray concentrations decline monotonically with altitude, consistent with the presence of local surface emissions from the strong winds."

Line 303-305: The authors note that the lack of vertical variability in dust concentration for ATom and E3SM is "consistent with the lack of local emissions from the underlying ocean surface". Do the authors not consider advection of dust from continental regions above the surface that may impact variability?

Reply: We have rephrased lines 330-335.

. "We find that the dust concentrations do not vary much from the surface up to 400 hPa in E3SM simulations and ATom measurements, which can be attributed to vertical mixing of the advected dust from the continental regions. In contrast, sea spray concentrations decline monotonically with altitude, consistent with the presence of local surface emissions driven by strong winds."

Figure 4: Do the horizontal line extensions (error bars) on each vertical point represent the range of values or variability in the ATom measurements? Please clarify this in the caption. Why is this not shown for the averaged model output as well?

Reply: We have included the standard deviation for both model and measurements in the revised version in Figure 4.

Line 311: Please clarify "good agreement in model vertical gradient". Is this assessed visually or statistically?

Reply: To provide additional information to support this statement, we have included the model standard deviation in Figure 4. We rephrased this sentence as follows:

Line 343-346: "Visual inspection of the simulated and observed vertical gradients in dust and sea salt aerosols show good agreement and therefore indicate that vertical mixing is likely a smaller concern, compared to other sources of biases in simulated INP concentrations in this region. The agreement between model and observations was assessed by checking if ATom observed mean values are within the simulated standard deviation for dust and sea salt.

"

Line 312: A source of literature here would provide support for this claim of microphysical/optical implication.

Reply: We have added the following references to support the claim of microphysical and optical implications.

Lines 347 -350

"The model evaluation of vertical aerosol profiles has implications for the role of INPs in cloud optical and microphysical properties in the MBL and free troposphere (Murray et al., 2021; Tan and Storelvmo, 2019; Burrows et al., 2022). Using lidar retrievals of dust, smoke, and cloud phase, Tan et al. (2014) showed that the presence of dust and smoke particles were negatively correlated with the supercooled liquid fraction."

Figure 6: The caption for this figure does not clarify what the acronym "FGE" means.

Reply: We have clarified FGE in the caption and added Equations and additional discussion for FGE in the text.

Figure 7: What are the values represented in the brackets of the legend? Are these values discussed in the text? This needs to be clarified in the caption.

Reply: We have clarified the values in the brackets in the caption for Figure 7.

Line 378-379: With very close visual inspection of Figure 7b, one could potentially see that CNT "agrees well with INP measurements", though it is a bit challenging due to the number of data points and numerous colors. Are the authors able to provide a statistical metric of agreement between the MICRE observed INP concentrations and those of the model values?

Reply : Although CNT appears to agree with INP measurements, CNT in E3SM has a compensating effect because the freezing rate is overestimated in the model and the dust aerosol concentrations in accumulation and coarse mode are underestimated in E3SM.

We discuss the potential reasons why CNT might agree well with INP measurements from MICRE in lines 464 - 466.

"Overestimation of dust immersion freezing rate coefficients in the Wang et al. (2014) parameterization used in E3SM (Cornwell et al., 2021), and 2) biases associated with using E3SMv1's CNT-based INP 10 s diagnostic as a proxy for INPs measured by the ice spectrometer. In order to test if this diagnostic is a good approximation for INP concentrations in Macquarie Island measured using an ice spectrometer, we perform a series of idealized droplet freezing experiments. "

We quantified how much the number of INPs predicted by E3SM's CNT parameterization would change if the parameterization were applied to an idealized ice spectrometer measurement performed at a constant cooling rate. We conducted idealized simulations of isothermal and constant cooling rate droplet freezing experiments to explore the implications of the time-dependent behavior assumed by the E3SM's CNT parameterization when compared with

observed INPs from the ice spectrometer. We perform sensitivity simulations for conditions representative of high and low INP regimes.

Line 410-415: Ship stack exhaust has been mentioned as a significant influence on aerosol and particle measurements during the MARCUS campaign (Humphries et al., 2021). Can the authors please provide a discussion on how this influence was removed from the dataset or how ship stack exhaust may influence/bias the MARCUS INP dataset?

Reply: We were mindful of ship stack influence and rated the sootiness or otherwise of the filters prior to processing. We did not find any significant ship stack influence standing out in the INP data in relation to the filter coloring from stack particles, which was in line with previously published investigations. Welti et al., 2020 analyzed soot and INP data from ship expeditions and found a moderate correlation between soot and INP number concentrations only at or below -36°C, much colder than the lower end of the MARCUS measurements. McCluskey et al., 2018 analyzed the relationship between black carbon measurements and INP concentrations from the CAPRICORN campaign in a similar region of the Southern Ocean, finding no correlation at any temperature >-26°C. Analyses to be published elsewhere also found no stack influence on total aerosol surface area in MARCUS, suggesting consistency with no INP influence.

Reference : Welti, A., Bigg, E. K., DeMott, P. J., Gong, X., Hartmann, M., Harvey, M., Henning, S., Herenz, P., Hill, T. C. J., Hornblow, B., Leck, C., Löffler, M., McCluskey, C. S., Rauker, A. M., Schmale, J., Tatzelt, C., van Pinxteren, M., and Stratmann, F.: Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern oceans, Atmos. Chem. Phys., 20, 15191–15206, https://doi.org/10.5194/acp-20-15191-2020, 2020.

McCluskey, C. S., Hill, T. C. J., Humphries, R. S., Rauker, A. M., Moreau, S., Strutton, P. G., ... & DeMott, P. J. (2018). Observations of ice nucleating particles over Southern Ocean waters. *Geophysical Research Letters*, *45*(21), 11-989.

Line 425-429: Are the authors able to point to literature that supports the claims of potential biases between MICRE INP and the model from island processes described in this passage?

Reply: We have revised this paragraph and added more references for impacts of island processes on INPs.

Lines 577 - 591

"Porter et al. (2022) found that small island sources off the coast of Russia contribute to high concentrations of biological INPs in the Arctic due to the nutrient rich water from the riverine sources. Their findings indicated that islands may be potential sources of biogenic INPs near the Russian coast. Although more work is needed to define the key sources of biological or biogenic INPs affecting MICRE INP concentrations, similar island processes may play a role in altering

the marine boundary- layer dynamics near Macquarie Island. These effects could potentially influence surface INP sources, losses, and boundary- layer mixing due to surface drag and orographic lifting caused by the island. Inoue et al. (2021) investigated the cruise data from the marginal ice zone in the Chukchi Sea and found high INP concentrations from sea salt and organic carbon above $-10^{\circ}C$ during the high wave conditions. Since MICRE collected samples near the surf zone, taller waves and high surface winds may increase sea spray supply to the INPs. Figure S5 shows that the sea spray aerosols emitted from wave breaking near the island regularly passes over the whole isthmus during the windy conditions regularly present at these latitudes before reaching the filter located on the island. The potential existence of local terrestrial or anthropogenic sources from the island could yield high INP concentrations during MICRE compared to open ocean INP samples from ACE or MARCUS campaigns in the SO. It would be interesting to employ a regional model with atmosphere-ocean-wave coupling to test these localized island processes. If local sources indeed dominate the INPs observed at MICRE, it may therefore be expected to be less directly comparable to global models than the open ocean measurements from ship-based campaigns such as MARCUS. "

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4. McCluskey, C. S., DeMott, P. J., Ma, P. L., & Burrows, S. M. (2019). Numerical representations of marine ice-nucleating particles in remote marine environments evaluated against observations. *Geophysical Research Letters*, *46*(13), 7838-7847.
5. McCluskey, C. S., DeMott, P. J., Prenni, A. J., Levin, E. J., McMeeking, G. R., Sullivan, A. P., ... &
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7. Schrod, J., Thomson, E. S., Weber, D., Kossmann, J., Pöhlker, C., Saturno, J., ... & Bingemer, H. G. (2020). Long-term deposition and condensation ice-nucleating particle measurements from four stations across the globe. *Atmospheric Chemistry and Physics*, *20*(24), 15983-16006.
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9. Tan, I., Barahona, D., & Coopman, Q. (2022). Potential Link Between Ice Nucleation and Climate Model Spread in Arctic Amplification. *Geophysical Research Letters*, *49*(4), e2021GL097373.
10. Vergara-Temprado, J., Miltenberger, A. K., Furtado, K., Grosvenor, D. P., Shipway, B. J., Hill, A. A., ... & Carslaw, K. S. (2018). Strong control of Southern Ocean cloud reflectivity by ice-nucleating particles. *Proceedings of the National Academy of Sciences*, *115*(11), 2687-2692.

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