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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

Referee comment on "Long-term variability in immersion-mode marine ice-nucleating particles from climate model simulations and observations" by Aishwarya Raman et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-203-RC1, 2022

General 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 parametrizations, 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.
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.

Specific comments:

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

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

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.

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

Line 101: quantify "temporally representative"

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

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

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

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.

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.

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.

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

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

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.

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.

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.

Line 313: Elaborate the implications of the model evaluation.

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.

Line 337: Clarify what the implications are.

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.

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

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

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

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

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

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.

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.

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.

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.

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

Table S1, line 519: should it be mineralogy?

Line 116-117: correct citation style

Line 213: Vali 1971 would be a better reference.

Line 284: …whereas it overestimates…

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

Line 289: correct citation style

Line 295: missing closing bracket

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

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

Table 3: M18 instead of M17

Line 387: correct citation style

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

Line 415: remove line break

Line 420: Jumping topic. Add line break.

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

Code availability: remove “also”

References:

Burrows, S. M., McCluskey, C. S., Cornwell, G., Steinke, I., Zhang, K., Zhao, B., et al. (2022). Ice-nucleating particles that impact clouds and climate: Observational and modeling research needs. Reviews of Geophysics, 60, e2021RG000745. https://doi.org/10.1029/2021RG000745

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Schrod, J., Thomson, E. S., Weber, D., Kossmann, J., Pöhlker, C., Saturno, J., Ditas, F., Artaxo, P., Clouard, V., Saurel, J.-M., Ebert, M., Curtius, J., and Bingemer, H. G.: Long-term deposition and condensation ice-nucleating particle measurements from four stations across the globe, Atmos. Chem. Phys., 20, 15983–16006, https://doi.org/10.5194/acp-20-15983-2020, 2020.

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