

Summary:

This study by Chatziparaschos et al. adds dust, terrestrial bioaerosols, and marine organic ice nucleating particles (INPs) to a global chemical-transport model (TM4-ECPL) and examines the contribution of each INP type at different latitudes, altitudes, and temperatures over simulations from 2009-2016. Dust INPs are found to dominate at higher altitudes ($<-20^{\circ}\text{C}$) across all latitudes and seasons. PBAP are the most important INP species at warm temperatures ($>-16^{\circ}\text{C}$), especially in terrestrial equatorial and mid-latitude regions, although the INP_{PBAP} parameterization appears to be overactive (Figure S6), so the exact contribution requires further study. Marine organics dominate the Southern Ocean at low altitudes in all seasons but have minimal contributions in the Northern Hemisphere. The results presented are in broad agreement with previous observational and modeling studies and provides a valuable addition to the INP literature.

The updated figures and restructuring of the discussion greatly enhance the manuscript and increase clarity for the reader. The manuscript now also includes a more detailed and nuanced discussion of potential model biases, which is appreciated and helps put the current study in context and provides suggestions for future modeling and observational studies. I have only one broader comment and some suggested minor (mainly grammatical errors and typos) edits.

Major Comments:

I still don't fully understand why 600 hPa was chosen as an example pressure level for Fig. 5. Although it is true that most INP measurements are made at a different temperature inside the instrument than the ambient temperature ($[\text{INP}]_{\text{T}}$ vs $[\text{INP}]_{\text{ambient}}$), the aerosol being measured is still more representative of the boundary layer than the free troposphere, just at a different temperature than ambient ($[\text{INP}]_{\text{T}}$). I agree that -20°C is a reasonable temperature to choose to be representative of MPC glaciation, but at high latitudes -20°C is reached within or very near to the boundary layer and the MPCs are typically not located in the free troposphere. Perhaps clarify that the pressure level chosen is representative of MPCs at low latitudes only, and not broadly representative of MPCs across all latitudes (lines 496-504). Much of the rest of the paper, and all the measurements being compared to in other figures are in the boundary layer, so it seems an odd choice to focus on the spatial distribution at a relatively high altitude, unless there is another reason I have missed.

The discussion of Fig. 5 is thorough, and the addition of the circled areas is helpful to follow the analyses. However, the mentions of “continental outflow” and “downwind of source areas”, etc would make more sense for a lower altitude than 600 hPa, either near the surface, or at least within the boundary layer. Extensive long range transport and mixing is expected for aerosols above the boundary layer, as well as a longer time since emission. There is no problem with discussing the results at this pressure/altitude, but trying to connect the results at 600 hPa to surface emissions perhaps needs to be more nuanced. This is particularly true since the high and low latitudes are discussed together, but the fixed altitude used does not account for the changes in boundary layer height (temperature) or vertical mixing that occur between the equatorial regions and poles.

Minor Comments:

1. Line 35-37: Suggest re-arranging the sentence as follows so it is easier to read: “MPOA-derived INP (INP_{MPOA}) prevails in the SH **at low altitudes**, particularly at subpolar and polar latitudes for temperatures...”
2. Line 40: Add “the” between “enhance” and “model’s”
3. Line 48: Suggest replacing “homogeneity of mixing ice” with “homogeneity **in** mixing **of** ice”
4. Line 64: “forms” should be “formed”
5. Line 66: Add “saturation” before “water vapor pressure”
6. Line 69: Remove “saturation” before “water vapor pressure” or replace with “**ambient** water vapor pressure are...”. This and minor comment #5 appear to be a mix-up in line numbers for a comment in the original review.
7. Line 73: “while it can also occur in cold clouds” seems out of place in a paragraph focused on MPCs.
8. Line 75: The sentence is a bit confusing, suggest replacing “air – can activate temperatures” with “air can activate at temperatures”.
9. Lines 94-97: Although it is true that INP prediction in models often uses empirical parameterizations, CNT (classical nucleation theory) is an alternative and can be used at temperatures above -35 °C. See Kanji et al. (2017) and references therein for a broad overview. A discussion of this topic is not required in this paper, and I suggest just removing the first half of the sentence and simply saying “INP prediction **in models** typically relies...”
10. Line 145: The acronym SIP typically refers to secondary ice **production**, not secondary ice particles as written in this line.
11. Line 149: What is meant by the “warmer parts of mid latitude atmosphere”?
12. Line 157: Replace “PBAP is critical contributor to INP.” with “PBAP are a critical contributor to INP concentrations”.
13. Line 182: Replace “this region” with “these regions”
14. Line 389: Suggest changing the title of Sec. 3.1.1 to “Global **aerosol** simulations” to separate them from the INP simulations presented later
15. Line 390: Add “multi-“ before “annual” for clarity
16. Line 397: Suggest replacing “high latitudes” with “**mid-high** latitudes” since MPOA seems to peak ~50°.
17. Lines 410-412: This sentence (“This comparison suggested...”) appears to have some typos and is hard to follow.
18. Line 423: Was “organic carbon derived from MPOA” meant to be “**MPOA** derived from **organic carbon**”?
19. Lines 431 and 441: Was “Austral Ocean” meant to be “**Southern** Ocean”?
20. Line 490: Suggest replacing “and are affecting the importance of various INP types” with “and affect the inferred importance of various INP types in the model”.
21. Line 500: No parentheses needed around the sentence “(This metric is explained...)”
22. Line 511: Consider adding “outside of strong dust source regions” after “40° latitude”, since it is clear that dust is a strong source in many terrestrial regions in the mid-latitudes.
23. Line 525: “isis” is a typo
24. Line 528: “shown in Figure 1) (Fig. 6a-d)” should be “shown in Figure 1) (Fig. 6a-c)”
25. Line 572: “mineral dust (Fig. 7d)” should be “mineral dust (Fig. **7a**)”
26. Line 572: “seasons (Fig. S3)” should be “seasons (Fig. **S4**)”

27. Line 591-592: Consider removing the sentence “The impact of...in the NH.” since it repeats the previous sentence.
28. Line 595: After “INP_{MPOA}”, consider referencing Fig. S4c, S5c, so the reader knows what figure you are discussing.
29. Line 598: After “INP increases”, consider referencing Fig. S4b, so the reader knows what figure you are discussing.
30. Line 657: I think the reference to “in summer (Fig. S2)” was meant to be “in summer (Fig. S4)”
31. Lines 689-691: Suggest clarifying the sentence as follows: “Since **only** a 20% overestimation of PBAP concentrations by the model can be deduced when compared to observations, an overestimation of the INP scheme seems to be the most plausible reason for the **large** INP_{PBAP} overestimation.”
32. Line 692: Does Fig. S6 compare the simulated INP types (INP_D, etc) against the total INP concentrations from observations? It does not appear that the observations have been separated by INP type. If not, suggest adding “total INP” between “INP types from” and “observations”.
33. Lines 706-712: Same as #32, does Fig. S7 compare the simulated INP types against the total INP concentrations from observations? If so, the good agreement between simulated INP_{PBAP} and total INP observations probably still indicates an overactive INP_{PBAP} parameterization, rather than good agreement with observations (which likely contain multiple INP types). The rest of the paragraph is great, and the points about local variability and the importance of bioaerosols at warm temperatures are important to make.
34. Section 3.3: The paragraph beginning with “Our results suggest that...” seems like a more logical place to begin this section, since it is more general and not specific to INP_{MPOA}. I would move that up to be the first paragraph of Sec. 3.3. I suggest removing the current first sentence (lines 714-716, “Earth System Models (ESM) encounter...” and moving the rest of the first paragraph (lines 716-720) “McCluskey et al. (2018) have reported...model’s underestimation of MPOA.” down to line 740 and combining it with the paragraph starting “Furthermore, INP dust parameterizations can introduce...other INP types on cloud properties”.
35. Line 728: “and used for global estimates” is unnecessary, since the rest of the sentence discusses their global applicability
36. Line 751: Was “ice nucleating ability” meant, rather than “dust nucleating ability”?
37. Lines 766-773: The paragraph “In summary, the comparison...account for INP_{PBAP} in climate modeling.” would fit better at the end of Sec. 3.2.3, or just remove since this information is covered in several places already.
38. Line 778: “singular description approach” should be “singular **hypothesis** approach”
39. Line 805: Suggest replacing “aerosol types as well as how” with “aerosol types **and** how”
40. Line 809: Suggest adding “on” between “cloud regimes and” and “climate”

Main Text Figure Notes:

1. Figure 3
 - a. Caption: there are two mentions of the units (ng-C m⁻³) and only one is necessary.
 - b. Fig. 3 also seems blurry, consider exporting in a different image format or increasing the resolution.
2. Fig 6 caption

- a. Square brackets are not needed around “[INP] number concentration”
 - b. The reference to panels (a-c) is confusing in “The colours show (a-c) the INP number concentration.” since the colors are the same in all panels (a-d) and not just (a-c).
3. Fig. 6 and 7 captions: Consider also specifying INP concentrations are only plotted if they exceed 0.01 m^{-3} , as in Fig. S4 and S5 captions.

SI Notes:

1. Figure S1
 - a. Check the legend on this figure, for example, the “ISAC_CNR_2012_Antarctica” and “Yin_China_2012” data have the same symbol, and the Antarctic data doesn’t appear to be plotted.
 - b. Are all the points marked as “Bigg_1969-1989” from Bigg’s papers, or are some from Welti et al. (2020) previously unpublished data (eg Tan1502, SHIPPO, etc)? I thought Bigg’s measurements largely ended $\sim 140^\circ\text{E}$ south of Australia. Also a note that Bigg’s Southern Ocean measurements are much higher (2-3 orders of magnitude) than all modern measurements (see e.g. McCluskey et al., 2018; Moore et al., 2024; Tatzelt et al., 2022) and may not provide the best comparison dataset for simulations in 2009-2016.
 - c. The reference to “Figure 4” was probably meant to be for Figure 10, and perhaps also Fig. S6 and S7.
2. Figure S6: The x-axis is plotted as Kelvin, but the label reads $^\circ\text{C}$ and the discussion of Fig. S6 in the main text refers to $^\circ\text{C}$ also. Please update to have consistent units.
3. Table S1: The “Bigg_1969-1989” data that was added to Fig. S1 does not appear to also have been added to Table S1.

References:

- Kanji, Z. A., Ladino, L. A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D. J., & Krämer, M. (2017). Overview of Ice Nucleating Particles. *Meteorological Monographs*, 58, 1.1-1.33. <https://doi.org/10.1175/AMSMONOGRAPHIS-D-16-0006.1>
- McCluskey, C. S., Hill, T. C. J., Humphries, R. S., Rauker, A. M., Moreau, S., Strutton, P. G., et al. (2018). Observations of Ice Nucleating Particles Over Southern Ocean Waters. *Geophysical Research Letters*, 45(21), 11,989-11,997. <https://doi.org/10.1029/2018GL079981>
- Moore, K. A., Hill, T. C. J., McCluskey, C. S., Twohy, C. H., Rainwater, B., Toohey, D. W., et al. (2024). Characterizing Ice Nucleating Particles Over the Southern Ocean Using Simultaneous Aircraft and Ship Observations. *Journal of Geophysical Research: Atmospheres*, 129(2), e2023JD039543. <https://doi.org/10.1029/2023JD039543>
- Tatzelt, C., Henning, S., Welti, A., Baccarini, A., Hartmann, M., Gysel-Beer, M., et al. (2022). Circum-Antarctic abundance and properties of CCN and INPs. *Atmospheric Chemistry and Physics*, 22(14), 9721–9745. <https://doi.org/10.5194/acp-22-9721-2022>
- Welti, A., Bigg, E. K., DeMott, P. J., Gong, X., Hartmann, M., Harvey, M., et al. (2020). Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern oceans. *Atmospheric Chemistry and Physics*, 20(23), 15191–15206. <https://doi.org/10.5194/acp-20-15191-2020>