REPLIES TO REVIEWER #2 COMMENTS

We thank the reviewer for the positive and constructive comments and the time they have spent in carefully reading our manuscript. All comments have been considered in the revised version of the manuscript. Below, in black are the reviewer comments and in blue our replies.

Summary:

This study by Chatziparaschos et al. examines the spatial and vertical distribution of ice nucleating particles (INPs) from three different sources: dust (D), marine organics (MPOA), and terrestrial bioaerosols (PBAP) through simulations using the TM4-ECPL global chemistry-transport model. They find dust are the dominant INPs at high altitudes/cold temperatures north of ~ 40°S, and across all seasons. PBAP are the most important INP species at warm temperatures (>-16 °C), especially in the Northern Hemisphere summer. Marine organic INPs are found to have minor contributions in the Northern Hemisphere, but to dominate in the Southern Hemisphere across all seasons. Overall, these results are in agreement with prior observational and modeling studies. The best agreement between simulated and observed INP concentrations across the entire temperature range (-5 to -35 °C) are found when both dust and MPOA INPs are included, but not PBAP. All simulations including PBAP INPs have a high bias relative to observations, which may point to an overactive parameterization for this species, which should be explored in future studies.

I found the article easy to read, and the figures are well-labeled and clear. However, the discussion needs to be re-structured into a more logical flow so the conclusions build on each other. This will help reduce some of the repetition of results between paragraphs and different sections. Overall, this work provides a valuable addition to the INP community, but some additional analyses in the comparison between simulated and observed INPs would be valuable and improve the manuscript greatly, so I am recommending minor revisions. Details are provided below:

We have restructured the manuscript, as also suggested by the other reviewer, and improved the discussion of model evaluation and uncertainties of our simulations and used this information for a thorough discussion of the results of the study. Our point-by-point replies to the comments follow in blue:

Major Comments:

1. The structure of the discussion is confusing, and lead to a lot of repetition of results, as well as mentioning results before the corresponding figure was presented. Additionally, the evaluation of the model against observations should come earlier in the discussion, since it impacts how the reader interprets the rest of the simulated results. A suggested order is listed below:
    1. Figure 1 (schematic representation of ice nucleation). This can also be removed, as it does not add much to the discussion and is only briefly mentioned. The two types of INP concentration metric indicated in the schematic are not well explained (see major comment #2).
    2. Presentation of the global model results (Figure 5)
    3. Comparison of the model against observations (Figure 6)
    4. Additional figures looking at specific aspects of the model (Fig. 2-4)

We have restructured the manuscript considering the suggestions of both reviewers.

We kept Figure 1 and added relevant discussion in the manuscript as provided in our reply to the next point of the reviewer.

Section 3 of the results now starts with the presentation of INP-relevant distributions (new Figures 2-4 see our reply to comment #1 of reviewer #1) and their evaluation focusing on PBAP, the evaluation of which was not shown earlier (new section 3.1- *INP-relevant aerosols*) (earlier point of the reviewer; new figures 3 and 4). Then, in section 3.2 the simulated global distributions are presented and discussed in 3 sub-sections:

section 3.2.1- *INP global distributions*

section 3.2.2- *Contribution of INP types to the total INP* where the percent contributions of each INP type to the total INP as deduced by the model are shown

section 3.2.3 -*Comparison of INP predictions with observations*

After this section we added a new section on:

"*3.3 Sources of uncertainties and implications for our results.*"

that collects the discussions on the uncertainties that in the initially submitted version were in different parts of the document. In such a way we avoid repetitions and we present the results of our study together with their confidence levels.

The abstract and the conclusions have been accordingly rephrased.

2. Figure 1 presents two different ways in which INP concentrations can be calculated/presented: the concentration which reaches a specific model level/altitude and are active at the model temperature, and the concentration at a specific model level which are active at a different temperature of interest. Since these numbers can be quite different, it would be helpful to the reader to have a description of what these values are and how they differ in the main text instead of just a very brief sentence in the caption for Figure 1. And then the INP metric plotted in each figure needs to be clarified in the main text and/or figure caption. I suspect the difference between these two metrics is driving the very different patterns seen between Fig. 3 and Fig. 4, for example, but this is not clear from either the text or figures.

Indeed, Figure 1 as well as $INP_{ambient}$ and INP_T needed explanation. Therefore, in the revised manuscript we added the following text at the end of the introduction.

"Figure 1 presents a comprehensive schematic illustration of INP formation derived from mineral dust, MPOA and PBAP. It includes emission processes, nucleation mechanisms, and the aerosol indirect effect, indicating the role of aerosols in cloud interactions and their impact on climate. ……

For this purpose, INP concentrations are presented in this study in two ways as $[INP]_{ambient}$ and as $[INP]_T$: $[INP]_{ambient}$ is calculated at ambient model temperature relevant to non-deep convective mixed-phase clouds using the ambient (model) temperature at each simulated level. $[INP]_T$ is calculated at a fixed temperature relevant to vertically extended clouds as deep-convective systems (Figure 1). $[INP]_{ambient}$ and $[INP]_T$ metrics can vary significantly."

later in Section 2.4 we also state:

"As outlined in the introduction $[INP]_T$ and $[INP]_{ambient}$ are determined by the simulated particle concentration and for $[INP]_T$ a specific temperature (T), while for $[INP]_{ambient}$ the model's ambient temperature is used. $[INP]_T$ is the appropriate metric when comparing modelled INP concentrations to observations, as measurements are typically conducted by exposing particles to specific, controlled

temperatures within the instruments. Thus, for the model evaluation, $[INP]_T$ concentrations are used that are calculated at the temperature at which the measurements were performed."

3. The model simulation of the aerosol types underlying the INP simulation, in this case, dust, sea spray, and PBAPs, are either only briefly mentioned as having previously occurred (dust, marine organics) or not covered at all (PBAPs). While reproducing these prior validation studies is unnecessary and impractical, I suggest adding a few sentences where the major conclusions, any biases, etc. of the previous evaluation studies (if any) are discussed. This would allow the reader to understand whether any of these biases may impact the INP results presented here.

All dust, marine organics simulations have been previously presented for specific years, without the INP component with the exception of dust that has been presented in Chatziparaschos et al., 2023. We thus focus the model evaluation on PBAP that has not been published earlier. A new section 3.1 has been added. In the sub-section 3.1.1 the global simulated distributions of the INP-relevant aerosols are presented (new Figure 2 provided in our replies to reviewer #1) and in sub-section 3.1.2 their evaluation by comparison with observations is presented (new figures 3, 4, S2 and S3 provided in our replies to reviewer #1). For dust, we summarize the outcome of the detailed evaluation done in our previous study (Chatziparaschos et al., 2023). For MPOA, in addition to reference to earlier studies, we show comparison of model results with clean air OC observations in the NH (Mace Head) and the SH (Amsterdam island). Finally, this session presents a detailed evaluation of PBAP simulations that has not been done in previous studies. Section 3.1.2 concludes :

'In summary, the evaluation of INP-relevant aerosols simulations shows that the model underpredicts MPOA (by about 30%; 3% - 50%) and overpredicts dust surface concentrations and deposition (by about 60%). PBAP concentrations are also slightly overpredicted (by about 20%), with BCT being significantly underpredicted and FNG moderately overpredicted. The model captures the seasonal variation of observed PBAP, while the simulated MPOA in the SH shows no significant variation across seasons. These model features must be taken into consideration when discussing the results of the INP simulations, since uncertainties in the INP simulations include uncertainties in both the INP parametrization and INP-relevant aerosol simulated concentrations and are affecting the importance of various INP types. '

4. The model validation against observations (Figure 6) could be improved in a few ways. First, based on Figure S1, the datasets used for evaluation are strongly biased towards the Northern Hemisphere, and the Arctic in particular. There is only one oceanic dataset included, and it occurs in a region where Saharan dust and possibly PBAP emissions are expected to dominate over marine INPs, so there are none or very few observations where MPOA INPs would be expected to be the dominant contribution. Oceanic INP measurements have been compiled in Welti et al. (2020), and many of them are from within the 2009-2016 period simulated here. Adding some of these to the evaluation would improve confidence in the simulated MPOA INP concentrations, in particular. More recent measurements of both PBAP and marine INPs exist as well, but would be more difficult to directly compare to the simulations due to the temporal offset. In addition, while the contribution of different model INP species are separated out and assessed, there appears to be no attempt to classify or separate the observations into either measured or expected dominant INP type. Why would you expect a PBAP-only parameterization to correctly simulate dust INP concentrations, for example? The fact that it is still overestimating concentrations when only PBAP are included (Fig. 6c) strongly suggests either the underlying PBAP aerosol emissions are too large and/or the INP parameterization is too active. Not all observations make measurements of INP species/type, but comparing the single-INP type simulations (Fig. 6a-c) to only observations where the INP type is known or can be reasonably estimated would improve confidence in the accuracy of the simulations. This

then has direct bearing on the percent contributions of each type calculated and shown in Fig. 2-4 and S2.

We thank the review for these comments and we tried to improve the simulated INP evaluation, further including information on dominant INP types for each observational data in addition to the evaluation of the INP-relevant aerosol in the new section 3.1.2.

All relevant to this study data from Welti et al. have been used.

Two new figures have been drawn to address the comment of the reviewer on 'separating the observations into either measured or expected dominant INP type'. The new Figure S7 shows the three different types of simulated INP and their comparison to the measured INP levels at each observational site for different latitudes zone in the atmosphere. This clearly shows the importance of different INP types for different latitude zones. For instance, in the NH extratropics (30N-60N) dust and terrestrial PBAP dominate INP levels, while in the SH extratropics (30S-60S) terrestrial (PBAP) and marine (MPOA) bioaerosol are dominating INP levels. In the tropics (30N-30S) dust and terrestrial aerosol appear the dominant types of INP.

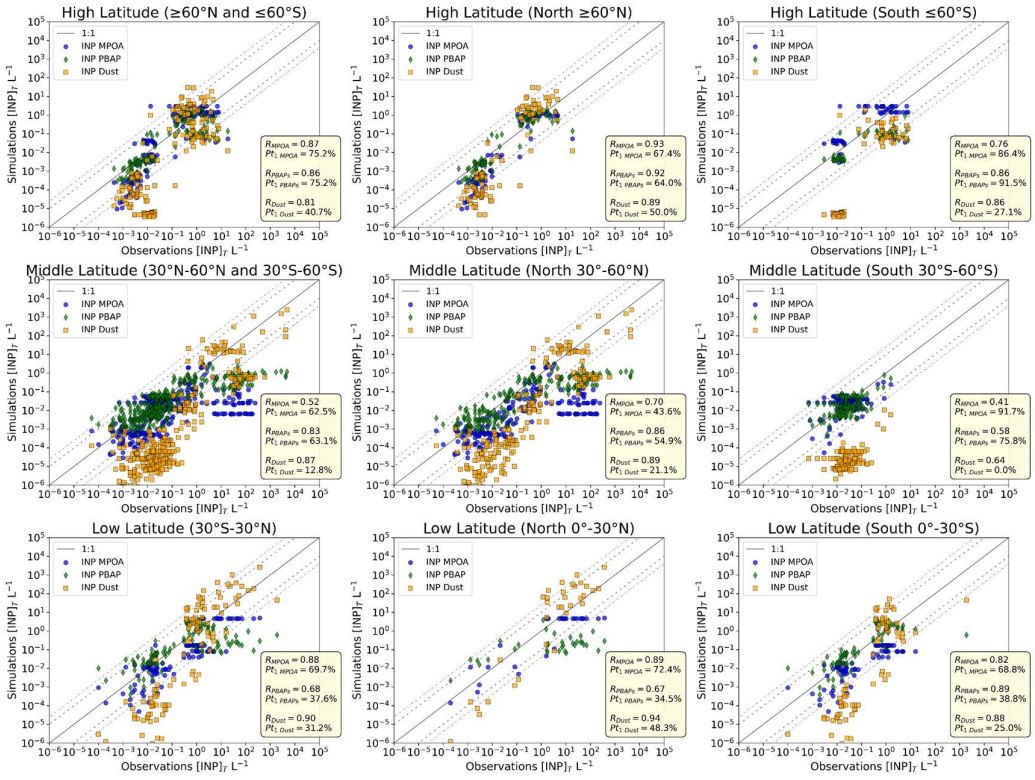

Figure S1: Comparison of INP concentrations calculated at the temperature of the measurements against observations accounting for mineral dust (orange), MPOA (blue), PBAP (green) separated in high, middle and low latitudes.

A second new Figure S6 has been added in the supplement that shows the difference between simulated INP for each of the aerosol types from the observed INP, all observational data included, classified by temperature bin from 230K to 275K (every 5 degrees). This figure demonstrates that the dust INP shows the smallest deviations from observations among the three INP types at the lowest temperatures (230K to 245K) and at warmer temperatures (255K to 270K) terrestrial PBAP and MPOA are performing better. For the 250K to 255K dust and terrestrial PBAP behave similarly.

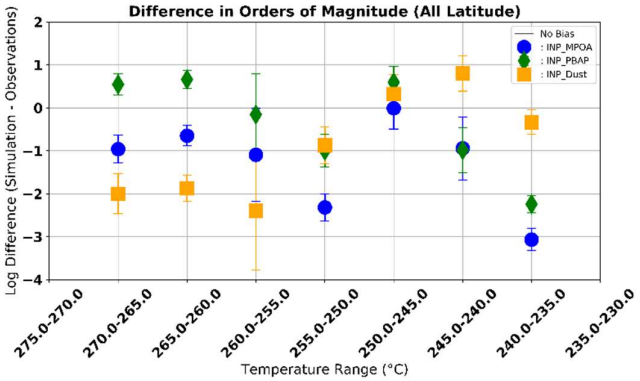

Figure S2: Order of magnitude / Logarithm of the deviation of simulated concentrations of INP types from total INP observations, as a function of binned temperatures. $INP_{\_Dust}$ in yellow, $INP_{\_PBAP}$ in green, $INP_{\_MPOA}$ in blue.

Relevant discussion is added in the revised manuscript.

Minor Comments:

1. Throughout the text, degree symbols are not always superscripted, and the 1 and 1.5 in Pt_1 and $Pt_{1.5}$ are not always subscripted.

   corrected

2. Throughout text: what is meant by INP "precursor". Based on context, it seems like INP "species" or "type" would be more appropriate and consistent with the language used by the community.

   Corrected throughout the manuscript

3. Line 26: replace ice "crystals" with ice "processes". DONE
4. Line 41: replace "dust and marine and" with "dust, marine, and" DONE
5. Line 42-44: the last sentence of the abstract does not fit with rest of the paragraph. I suggest just removing it. DONE
6. Line 49: define mixed-phase clouds as MPC here, otherwise you do not define the acronym before you use it. DONE
7. Line 50-51: The sentence "A significant feature…" is confusing. What do you mean by this/why is it important?

   The sentence is now rephrased: 'A significant feature of MPC is the degree of homogeneity of mixing ice particles and liquid droplets that affects the rate of precipitation formation (Korolev and Milbrandt, 2022)'

8. Line 60: remove "saturation" from "saturation water vapor pressure". You appear to be discussing factors that alter ambient vapor pressure and not the saturation value, which is an intrinsic value characteristic of a set of environmental conditions.

   DONE

9. Line 61: add “area” after “ice crystal surface” DONE
10. Line 69-71: Please include references to support that SIP influences “precipitation ranges, cloud properties, and albedo.” What are precipitation ranges?

    References are included. ‘Precipitation ranges’ has been removed.

11. Line 117: “INP contributor” should be “INP contribution” DONE
12. Line 117: “Nevertheless, there are other minerals” should be changed to something like “There are also other minerals” DONE
13. Line 131-132: “Recent advances in remote sensing and model/sensor fusion have enabled the presence of secondary ice in clouds, revealing that this process holds…” should be changed to “Recent advances in remote sensing and model/sensor fusion have enabled **detection** of secondary ice **processes** in clouds, revealing that **they hold**…”

    corrected

14. Line 137: “,with about” should be “, accounting for” DONE
15. Line 149: typo in “sea spay”, should be “sea spray” DONE
16. Line 152: Burrows et al. (2013) and DeMott et al. (2016), in addition to Wilson et al. (2015) are more appropriate references instead of Mitts et al. (2021), as they are some of the first studies to suggest this idea.

corrected

17. Line 153: MPOA and SS should be reversed in this sentence.

DONE

18. Line 167-170: This entire sentence is almost word for word copied from Wilson et al. (2015). Either indicate it is a direct quotation, or rephrase in your own words.

We thank the reviewer for pointing this out. The sentence is now rephrased and reads as follows:’ Marine INP concentrations are found to maximize over areas of high biological activity and wind speed in the Southern Oceans, the North Atlantic, and the North Pacific, while mineral dust from deserts (the main terrestrial source of INP) accounts for only a small fraction of the INP observed in this region (Wilson et al., 2015; Huang et al., 2018)’

19. Line 179-180: This sentence is a bit misleading. The simulations are based on others’ work identifying major INP types, which you (reasonably) take advantage of. Please rephrase to more accurately describe your contribution.

Presentation and discussion of Figure 1 has been changed and extended as stated in our reply to the major comment #2 of the reviewer.

20. Lines 239-241: How representative are “diverse locations across Europe” for PBAP emissions globally? Especially since Fig. 5 indicates the PBAP INP simulations peak in Africa and South America? Can you comment on how representative the Global Land Cover database from 2000 is for simulations that occur 1-2 decades later?

Indeed, these are very good points raised by the reviewer. Parameterizations based on observations over Europe might not be accurate for the flux calculations over other regions of the globe. This is now commented in the manuscript.” This parameterization is informed by field measurements of fluorescent biological aerosol particles conducted at diverse locations across Europe; thus, it might not be very

accurate for other regions and will be improved when data from other regions of the globe will become available."

Copernicus provides now Land Cover data at very high spatial resolution for the years 2015-2019 updated every year. Note that our simulations cover the years 2009-2016 and have to be considered as a climatological view of the atmosphere over that period. Thus, using a 2000 Global Land Cover database introduces some uncertainties but is not expected to qualitatively change the conclusions of the study. In the text we have added : "which represents a climatology for the present study since no changes in vegetation from year-to year are taken into account"

21. Line 269: Are MPOA and SS internally or externally mixed in the model? This will impact what diameters are simulated.

MPOA is considered to be a coating of SS thus internally mixed as described in detail in O'Dowd et al. (2008) and Vignati et al. (2010). To avoid any confusion we rephrased this sentence as follows: MPOA is emitted in the fine mode together with SS in amount that depends on Chl-a in the seawater and assuming that it is entirely insoluble as determined by O'Dowd et al. (2008) and applying the same modeling approach as in (Vignati et al., 2010).

22. Line 288: What is meant by "the spectrum of ice nucleation properties"?

corrected to 'ice nucleation spectrum'

23. Line 317-318: The sentence "Additionally, observations…" does not fit well in this paragraph.

We moved this sentence one sentence up. The text now reads" The number fractions of FBAP to the total aerosol particles in the super-micron size range could be relatively similar to those of PBAP under dry conditions (Tobo et al., 2013). Indeed, observations by Negron et al. (2020) suggest that airborne bacteria may be unambiguously detected with autofluorescence. However, during and after rain events, PBAP were underestimated by more than a factor of 2 (Huffman et al., 2013), suggesting that FBAP reflect only a portion of PBAP."

24. Line 330: In equation 3, is C_{MPOA} meant to be C_{TOC}?

corrected

25. Line 340: Simulated INP concentrations were compared to measurements at the same temperature and date. Were the lat/lon and altitude of the measurements also matched?

Yes. Text now reads: 'All model results are compared to observations for the specific month and year as well as location of observation, except for those reported by Yin et al. (2012) and Bigg campaigns (Bigg, 1990, 1973), which cover temporally scattered measurements (between 1963 and 2003) and are therefore compared to the modelled multi-annual monthly mean INP concentration from 2009 to 2016.'

26. Paragraph beginning with line 347: This paragraph jumps around and is hard to follow. The altitudes, temperatures, latitudes, and INP type discussed vary from sentence to sentence. It would be helpful to discuss Figure 2 for all INP types, then move on to figure S2 and the seasonality of the simulations, instead of sprinkling that into some of the paragraphs and then repeating much of that information later in the discussion. Focus this first paragraph on dust and move the couple of sentences about PBAP and MPOA to the next 2 paragraphs.

This section has been reorganized following the reviewer's suggestion.

27. Line 358: “amount of minerals” should be changed to something like “type and fraction of different minerals”

Change performed

28. Line 361-362: The statement that “PBAP are the primary source of INP between −12°C and −20°C.” should come after the evaluation of model results demonstrating the overestimation of PBAP by the model. See major comment #1.

In response to the reviewer comment #1 we have added a new section 3.1 evaluating PBAP simulations that has shown an overestimate with a normalized mean bias (NMB) for the total PBAP of 17% and overestimate (NMB 7%) for fungal spores and underestimate (-75%) for bacteria. This evaluation section is now before the discussion of the INP results.

29. Line 372-373: The sentence “INP_{PBAP} mainly affect…” simply repeats what was already said and is unnecessary.

The sentence has been removed.

30. Line 374: “(Fig. 2S)” is actually Fig. S2.

corrected

31. Paragraph beginning with line 369 needs some work, which will be helped by restructuring the discussion in line with major comment #1. The sentence beginning with “In boreal summer…” belongs with a discussion of Fig. S2 and not Fig. 2, and the one beginning “Our results suggest that PBAP…” with the discussion of Fig. 5. The last two sentences of this paragraph jump around between INP types and repeat things that were said in the preceding paragraph.

Corrections done as suggested

32. Line 383: Clarify that the “concentration range **shown in Wilson et al. (2015)…**” are, in fact, simulated concentrations and not actual observations.

For clarity, we rephrased as ‘simulated concentration range shown in Wilson et al. (2015)’

33. Line 387: Chubb et al. (2013) presented measurements of SLW and not INPs. Either clarify this, or cite something specific to INPs, such as McCluskey, Hill, et al. (2018) or Welti et al. (2020).

We now refer to McCluskey et al. (2018) and Welti et al. (2020)

34. Line 391: What models or types or models (ie global, cloud resolving) are you referring to? There have been significant improvements in many models between CMIP5 and CMIP6, also.

We replaced models by ‘Earth System Models’

35. Line 395: See Gettelman et al. (2020) and Bodas-Salcedo et al. (2019) for improvements in radiation budgets following an increase in SLW in two different models, in contrast to Huang et al. (2018).

This part of the discussion has been removed during the restructuring of the manuscript.

36. Line 395-397: See also McCluskey et al. (2023), which shows significant differences in CAM6 between two dust parameterizations, one of which was used in Huang et al. (2018) and overpredicts dust INP in the Southern Ocean, at least. I am not familiar with the validation of the ECHAM6-HAM2 model used in Huang et al. (2018), but simulated dust concentrations will also strongly impact these results because of the large discrepancy in freezing efficiency between marine and dust aerosol.

We have included the reviewer's comment in the manuscript – in the new section 3.3. on uncertainties we now refer to this source of uncertainty in model simulations:

" INP dust parameterizations can introduce significant biases in the calculations: INP_D simulations by CAM6 model were found to vary by a factor of 2 at temperatures lower than −25°C when two different parameterizations were used (McCluskey et al., 2023) with the one overpredicting INP from dust aerosol. Given the significant differences in freezing efficiency between marine and dust aerosols, simulated dust concentrations play a critical role in influencing study results, particularly in marine regions. An overestimation of INP_D could mask the potential effect of other INP types on cloud properties. "

37. Line 402: "figure 2c" should be capitalized. Also, it is not obvious in Fig. 2c that MPOA are enhanced at the surface, especially not in the Arctic. Please specify exactly what is plotted in Fig. 2- are the concentrations showing only INP active at the model temperature level, such that MPOA do not appear enhanced at the surface because they are not active at very warm temperatures? Please also check the colorbar scale for Fig. 2d-f. Based on Fig. S2, I would expect the MPOA % contribution to be higher in Fig. 2f, since it dominates from 60-90 °S and up to ~500 hPa during all seasons.

The old Figure 2, in the revised manuscript is split in two new Figures 6 and 7 that are also redrawn for better quality. Indeed, these figures (Figure 6) shows only active INP number concentrations at ambient (model's) temperature and the scale is in L^{-1}. Figures 7 show fractional contributions of the different types of INP to the total simulated INP and the scale is in percent units. These are computed for ambient (model) temperatures.

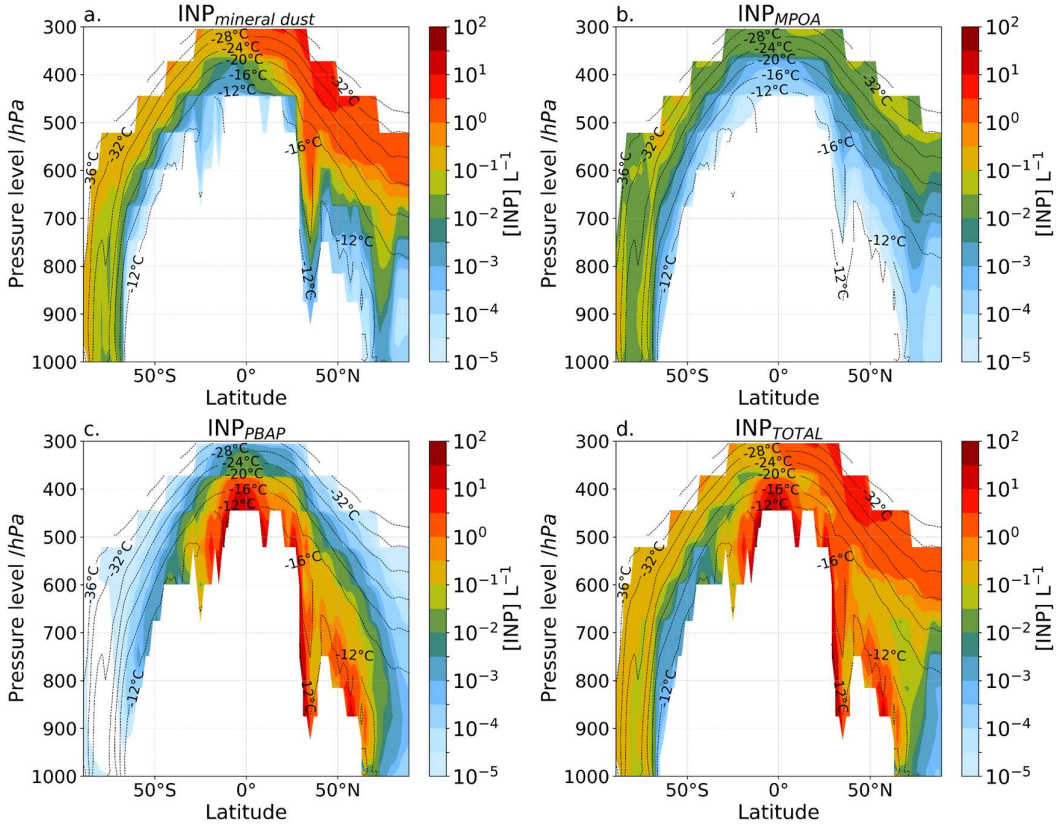

Figure 6: Multi-annual averaged zonal mean profiles of [INP] number concentration calculated at modelled ambient temperature by TM4-ECPL and accounting for (a) mineral dust, (b) marine bioaerosols, (c) fungal spores and bacteria (PBAP) and (d)all INP types in the model. The black contour dashed lines show the annual mean temperature of the model. The colours show (a-c) the INP number concentration.

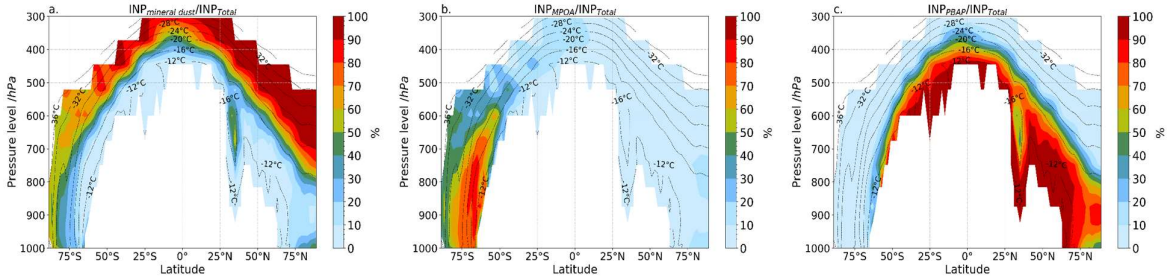

Figure 7: Multi-annual averaged zonal mean profiles of the percentage contribution of each species to the total INP number concentration, calculated at modelled ambient temperature by TM4-ECPL, showing contributions from (a) mineral dust, (b) marine bioaerosols and (c) fungal spores and bacteria (PBAP). Black dashed contour lines represent the model's annual mean temperature.

38. Line 404: Replace “high pressure levels” with “warmer temperatures” for clarity.

done

39. Line 407-410: Note Fig. S3 in addition to S2, otherwise it is not referenced anywhere in the main text. The statement about PBAP concentrations refers to S3, for example, but is not listed. This location may change if the discussion is re-structured.

done

40. Line 411-412: PBAP INPs appear to dominate at low altitudes in the Northern Hemisphere year-round, based on S2, and likewise for MPOA INPs in the Southern Hemisphere. The seasonality is not clear in Fig. S2, except for >60 °N. Also, what is meant by "showing alternating patterns influenced by vegetation and ocean biota"?

This discussion has been modified:

"In the boreal summer, the high INP_{PBAP} contributions to total INP are shifted to northern latitudes due to vegetation growth (Fig. S4). … There is a pronounced seasonality in dust INP (Figure S5), with large concentrations observed between 40°N and 90°N during the boreal winter and spring (DJF, MAM) when transported dust can influence INP concentration over Arctic. In contrast, INP_{MPOA} shows minimal seasonal variation and consistently dominates the SH between 40°S and 80°S. Between 60°S and 60°N, INP_{PBAP} is the most prevalent INP type at higher pressure and temperature levels, especially in the NH during the boreal summer (JJA) and autumn (SON), when its contribution to total INP increases."

41. Line 415-417: It is a little difficult to directly compare Fig. S2 and S3 to observational data which was all made at the surface. The concentration of marine INPs active at any given temperature would be expected to be higher near the surface, since that's the source. But since I think you have plotted the number contributing at a seasonally averaged isotherm, your percentages will be very sensitive to the exact parameterization used to determine the temperature dependence. Please clarify if what is being plotted is the INP concentration at the model temperature and altitude. If not, have you explored why the MPOA and PBAP do not appear to peak at the surface? You should also clarify that the region you are discussing here is the Arctic (ie >60 °N), and the altitude/temperature range you are referring to. Could the fact that PBAP appear to contribute more than MPOA in summer be related to the apparent overestimation of INP PBAP by the model (Fig. 6)?

For PBAP and MPOA to act as INP since for PBAP the temperatures should be in the range of [-34°C, -9°C] and for MPOA in the range of [-27°C, -6°C] and for dust k-feldspar in the range of [-37.5°C, -3.5°C] and quartz in the range of [-37.5°C, -10.5°C] as was described in section 2.3. Furthermore, the contribution of the INP types (Figure 7, initially Figure 2d,e,f, and Figures S4, S5, initially Figures S2 and S3) to the total INP depends on both the ambient temperature and on the actual concentrations of all three particle types PBAP, MPOA and dust, whereas the INP_{MPOA} and its seasonality (Figures S4, S5) depend on the temperature and the actual concentrations of only MPOA which are expected to vary seasonally. The larger contribution of PBAP to INP than MPOA at >60°N in summer could be due to an overestimate of PBAP levels as discussed in new section 3.1.2 on model evaluation.

This is now stated in the manuscript "This discrepancy may be due to the underprediction of MPOA and overprediction of PBAP and dust concentrations discussed in section 3.1.2."

The sensitivity of the results to the INP parameterizations used is discussed in the new section 3.3. on uncertainties.

Furthermore, the figure captions now clarify that the black contour lines represent seasonal (annual) mean isotherms in degrees centigrade.

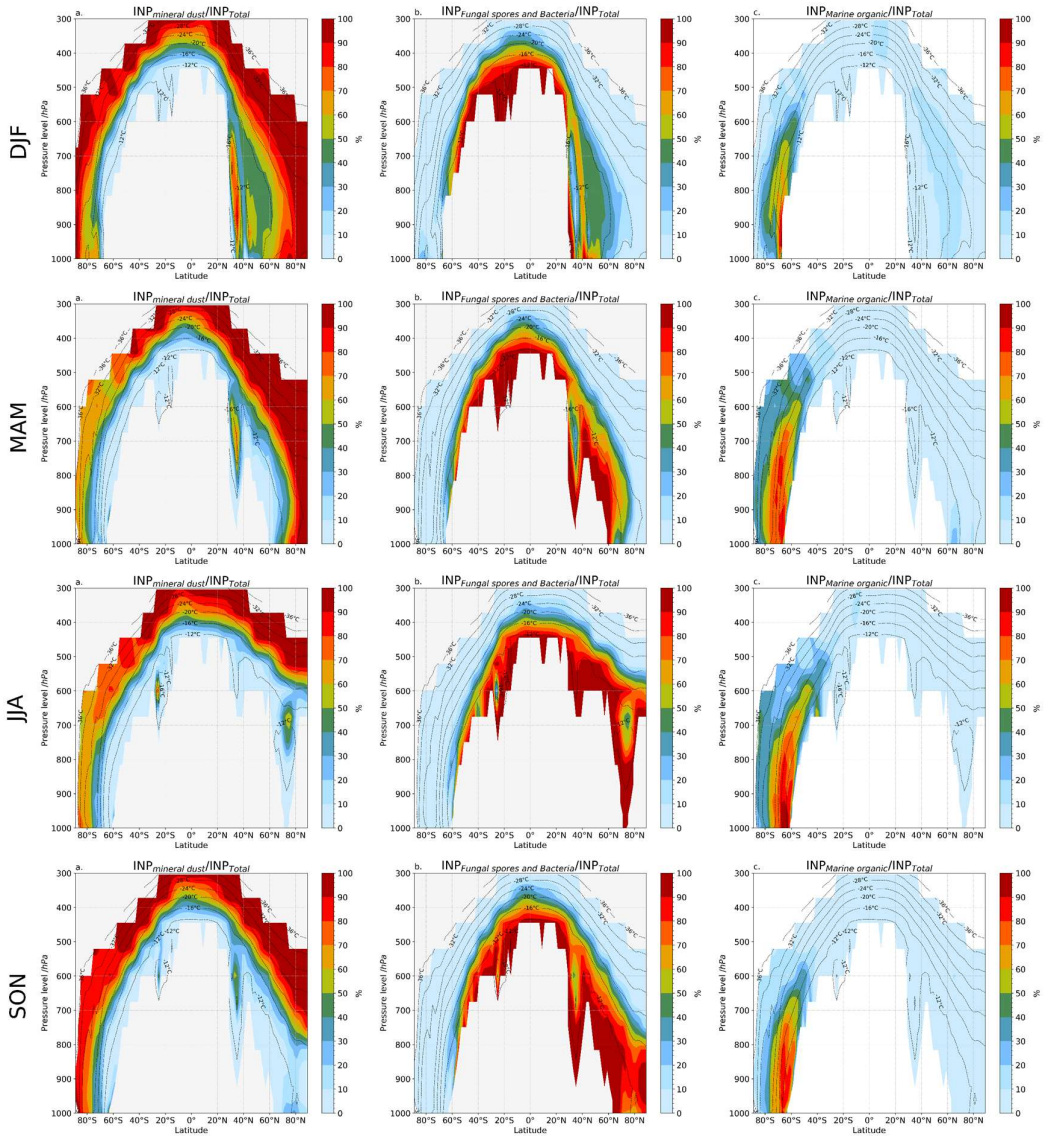

Figure S3: This figure depicts the seasonal percentage contribution of a) mineral dust, (b) fungal spores and bacteria and (c) marine organic aerosols calculated by TM4-ECPL where the total $[INP]_{ambient}$ concentration is larger than $0.01 m^{-3}$. The black contour lines represent seasonal mean isotherms in degrees centigrade.

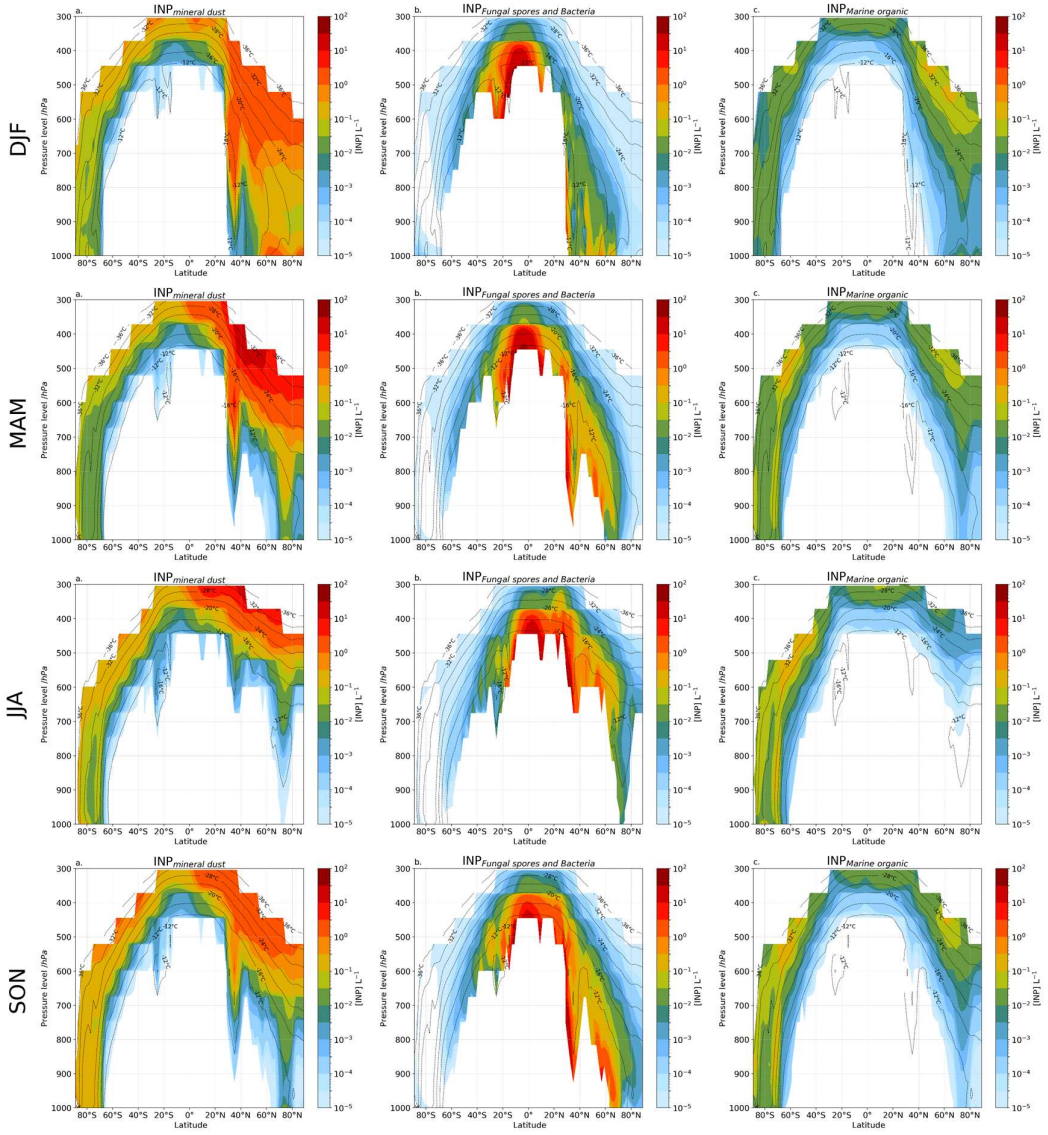

Figure S4: This figure depicts the seasonal concentration of a) mineral dust, (b) fungal spores and bacteria and (c) marine organic aerosols calculated by TM4-ECPL where the total $[INP]_{ambient}$ concentration is larger than $0.01 m^{-3}$. The black contour lines represent seasonal mean isotherms in degrees centigrade.

42. Line 431-432: Capitalize F in figure 3, replace "INP total number concentration column" with "total INP column (number) concentration". Can you also clarify what INP metric is plotted in Fig. 3? Is it the concentration of INPs active at each temperature which reach the range of temperatures listed (ie the -40 to -30°C values are at higher altitudes and the -10 to -20°C values are from near the surface) or the "potential" total concentration active at each temperature, regardless of whether they reach the altitude corresponding to the listed temperature range?

This figure (Figure 3 in the initial submission) shows the total INP column (up to 300 hPa) calculated by sampling the model based on the model temperature that is separated in three distinct temperature bins [-40 °C, -30 °C], [-30 °C, -20 °C] and [-20 °C, -10°C]. So, it shows the actual (not the potential) total INP active at each temperature bin in the model column up to 300 hPa. This figure has been replaced by a new one (Figure 9 in the revised manuscript) that shows each type of INP separately at the three ranges of temperature. This is now clarified in the manuscript:

"To further illustrate the different components of INP depending on atmospheric temperature, Figure 9 presents the percent contributions of INP types calculated at modelled ambient temperature and classified in three temperature ranges: [-40°C, -30°C], [-30°C, -20°C], and [-20°C, -10°C]. The figure highlights the spatial variability as a function of temperature of the contribution of the studied species to the total simulated INP at modelled ambient temperature. In order to plot this figure, all model results for which modelled temperature falls into the respective temperature range are selected. The percent contributions of each INP type to the total INP in each of the model grids and instances are calculated and then averaged per model grid for all altitudes. "

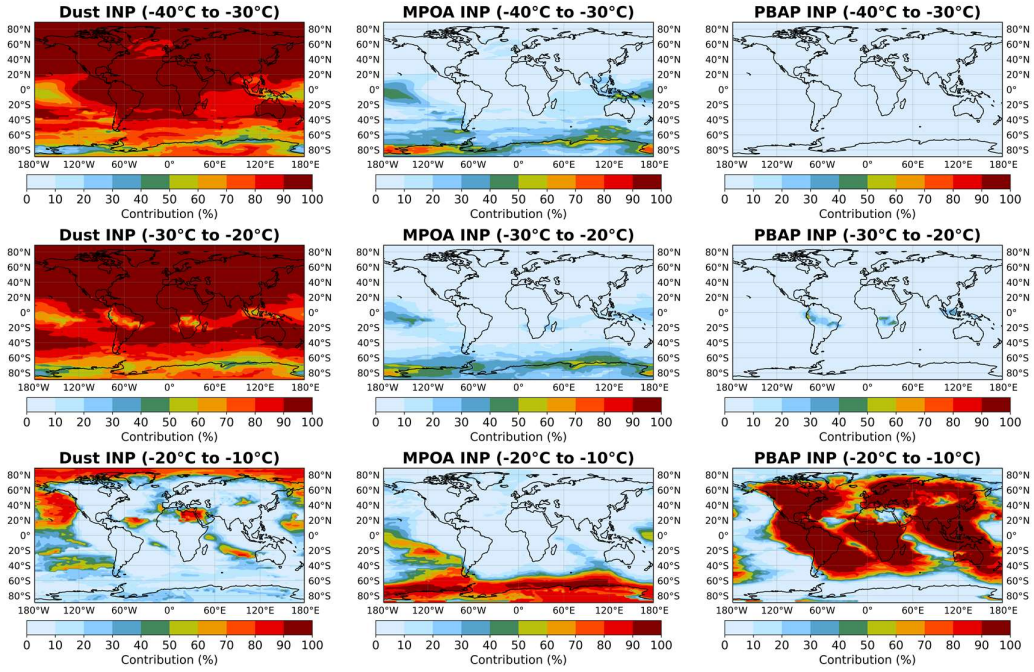

Figure 9: Multi-annual averaged percentage contributions of INP sources to the total INP column (surface up to 300 hPa): INP from mineral dust (left column), INP from MPOA (middle column), and INP from PBAP (right column), categorized by modelled temperature ranges: (top row) -40°C to -30°C, (middle row) -30°C to -20°C, and (bottom row) -20°C to -10°C. INP are calculated at modelled ambient temperature.

43. Line 434: Is the annual mean % contribution listed also from 2015? How much annual variability is simulated by the model?

All figures now refer to multi-year simulation results.

44. Line 444: "INP sources" is more correctly "INP types" or "INP species"

Corrected throughout the text to INP types

45. Line 444-445: Please clarify what is meant by "correlation between the percentage contribution of each INP type and the concentration ranges of INP." What is a concentration range? The % contribution is calculated from the concentration of one species divided by the total, so must mathematically be correlated with the species concentration.

For clarity this figure has been replaced by a new figure number 8 and discussion has been modified accordingly

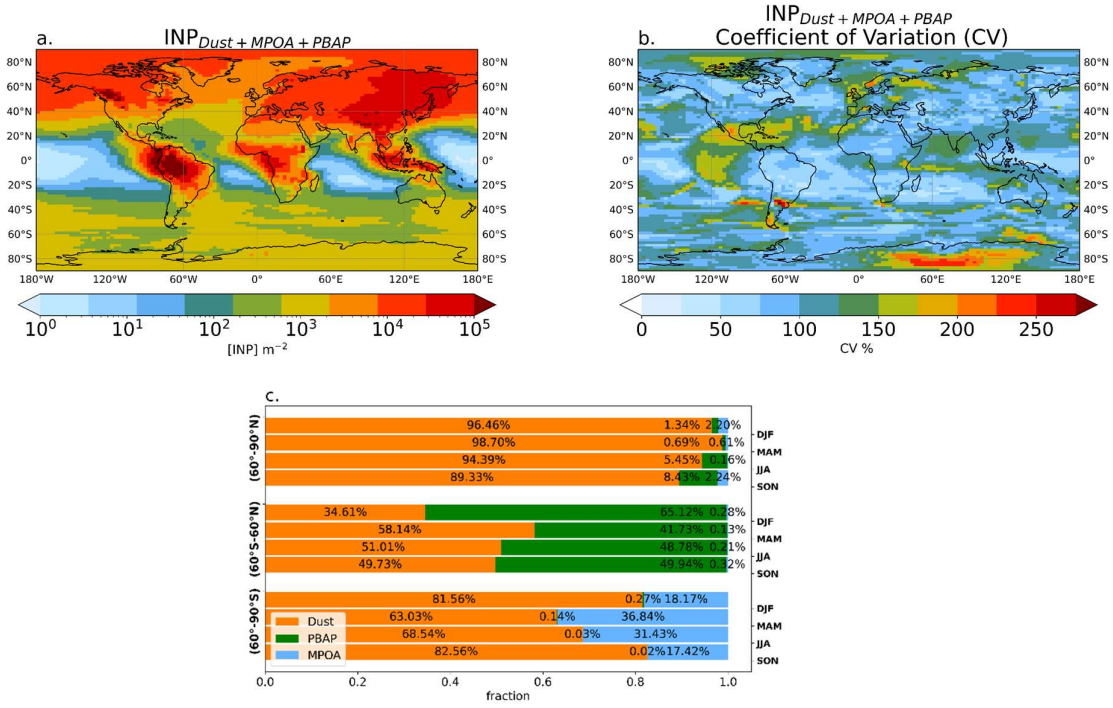

Figure 8: (a) Annual mean INP column number concentrations up to 300 hPa, for INP at modelled ambient temperature,(b) the interannual variability expressed by the coefficient of variation that is calculated as the standard deviation of the annual mean columns to the multiyear annual mean and is expressed in percent and (c) seasonal percentage contributions of INP from mineral dust (orange), MPOA (light blue), and PBAP (green), with data separated by middle latitudes (60°S–60°N) and high latitudes (60°–90°N, 60°–90°S).

46. Line 457-458: Is 27% the annual average of INP_{PBAP}, if so, for which year? The fall INP_{PBAP} contribution is 45%, but it is lower in the summer, ~30%. Provide an average if you wish to talk about both together.

Following the change of figure to new Figure 8, the contribution of the different INP types is now discussed on a latitudinal zone and seasonal basis. This discussion now reads:

'Figure 8 provides a comprehensive view of the global distribution of INP in the tropospheric column from the surface up to 300 hPa (Fig. 8a), calculated at the ambient modelled temperature, as well as the percentage contribution of each INP type per season and for three latitudinal zones (Fig. 8c). High INP column burdens are simulated in the NH and especially over the Sahara, parts of the Middle East, and the northern part of South America. These regions are known to be significant dust emission and biological aerosol sources (Amazon). Lower INP column burdens are simulated over the polar and oceanic regions, indicating that the contributions from INP sources in these areas are limited. The simulated INP column burdens are greater in the NH than in the SH, which is in agreement with previous studies (Vergara-Temprado et al., 2017). Figure 8b shows the associated interannual variability of the simulated INP multiyear mean columns indicated by the coefficient of variation that is calculated as the standard deviation of the annual mean columns to the multiyear annual mean and is expressed in percent. We find that locally the interannual variability can exceed 150%, particularly in the tropics and the polar regions and has to be kept in mind when interpreting modeling results. Figure 8c shows that mineral dust is the dominant source of INP in the high northern latitudes (60°-90°N) across all

seasons, with contributions exceeding 89% in every season. INP_{PBAP} and INP_{MPOA} play minimal roles, with INP_{PBAP} contribution to the column burden of the total INP peaking at about 8.5% in the fall (SON). In mid-latitudes and the tropics (60°S-60°N), INP contributions are more seasonally variable, highlighting the interplay between dust and biological particles in this region. Mineral dust dominates during MAM (spring NH) and SON (autumn NH), while INP_{PBAP} plays a prominent role in DJF (winter NH), contributing about 65%. In the high southern latitudes (60-90°S), mineral dust remains the main source of INP, especially in DJF (summer SH), where it accounts for about 82.5%. However, in contrast to the other latitude zones, INP_{MPOA} shows significant contributions to the total INP column burden in MAM (about 37%) and JJA (about 31.5%) in this latitude zone, while INP_{PBAP} contribution is negligible.'

47. Line 466 and 472: Do you mean the spatial "distributions of INPs"? Either clarify or just replace with concentration.

This now reads:' the simulated spatial distributions of INP concentrations'

48. Line 467: Why was 600 hPa chosen as an example pressure level? Most of the observations you compare to later (Table S1) appear to be from ground sites, so surface concentrations at any temperature of interest would seem more appropriate. Also, Fig, 4a appears to show INP concentrations below detection limit between ~30 °S and 30 °N at 600hPa. How do you have almost global coverage in Fig. 3 and 5? It is not clear in most of the figures which metric of INP concentration you are plotting, and it is difficult to decipher why there are large differences between the maps.

The pressure of 600 hPa corresponds on average to about 4 km i.e. is above the boundary layer in the free troposphere. The INP samples even from near- ground have been measured at instrument's temperature which is much lower than the ambient one. Accounting for an adiabatic dry air temperature decrease of 6.5 °C/km (about 6°C/km for wet air), at 4km height temperature should be 24-26°C lower than at surface. For an average surface temperature of about 14°C, this ends up to about -14 °C to -16°C average temperature at 4km, which is close to the instrument's temperature when measuring INP.

The following explanation is added in the revised manuscript: "This pressure level corresponds to the low free troposphere and average temperatures broadly consistent those used for the INP measurements."

49. Line 468-469: What is meant by "allow all species to activate and act as INP"?

The sentence now reads. "These conditions of temperature and pressure are representative of MPC's glaciation."

50. Paragraph beginning with line 474: You don't really explain what is meant by INP_{D-20} or $INP_{PBAP-20}$, etc, consider replacing with, eg, "INP_D at -20 °C".

Modified as suggested throughout the manuscript.

51. Lines 475-476: The phrasing "transport patterns from sources that favour dust present in the NH." is confusing. The wording used in the conclusion is clearer.

This sentence has been rephrased as follows: "[INP_D[600hPa, -20°C] (Fig. 5a) are higher in the mid-latitudes of the NH than in the SH due to the location of dust sources and long-range atmospheric transport patterns that favour the presence of atmospheric dust in the NH."

52. Line 482: How is the upper troposphere a "marine environment"? In addition to the possible influence of PBAP in marine environments, dust INPs contribute significantly in many areas. This is particularly clear over the central Atlantic, downwind of the Sahara, where dust INP concentrations are larger than both MPOA and PBAP. And dust also contributes significantly over the N Pacific down wind of continental outflow.

This sentence has been rephrased for clarity: "Remarkably, there are oceanic regions in the South Atlantic Ocean and Pacific Ocean (such as the south hemisphere tropical west coasts of South Africa and South America) where PBAP has the potential to form ice crystals at the outflow of the continental air (Fig. 5d, circles 1 and 5), enhancing INP concentrations in marine atmosphere." Further in the discussion of that figure we also comment: "INP_D[600hPa, -20°C] shows also significant levels over the North Pacific, where dust is carried by continental outflows. Mineral dust and MPOA control the INP population over South Atlantic adjacent to North east coast of S. America due to Patagonia desert and marine biota (Fig. 5d, circle 3), while over southern Indian Ocean INP_{MPOA}[600hPa, -20°C] dominates (Fig. 5d, circle 4)."

53. Line 484: It would be appropriate to add somewhere a discussion of the observations, and the mix of INP types observed or expected to be observed in those locations. There are no observations included, for example, where MPOA are expected to be the dominant INP type, since the only cruise included is immediately down wind of Africa, where significant dust and PBAP emissions are expected. See Major Comment #4.

In the revised version of the manuscript, a new supplementary figure S7 has been added which, using the model results, separates the observations into the expected dominant INP type. See our response to major point 4 for further details.

54. Line 488-489: What altitude/pressure were the simulated concentrations in Fig. 6? If the 600 hPa from Fig. 5, why, when the majority of observations are surface measurements? Did the lat/lon and altitude of the simulated values match each observation?

Comparison is done on lat/lon/altitude basis of the observations and NOT at 600 hPa. We also reported this in our reply to point 25 of the reviewer: 'All model results are compared to observations for the specific month and year as well as location of observation'

55. Line 495: Simulating dust INPs alone appears to compare quite well against the observations below ~-25 °C.

Yes, this is now demonstrated better with the new supplementary figure S6 that shows the difference between simulated INP for each of the aerosol types from the observed INP, all observational data included, classified by temperature bin from 230K to 275K (every 5 degrees). This figure demonstrates that the dust INP shows the smallest deviations from observations among the three INP types at the lowest temperatures (230K to 245K) and at warmer temperatures (255K to 270K) terrestrial PBAP and MPOA are performing better. For the 250K to 255K temperatures dust and terrestrial PBAP behave similarly.

56. Line 501: "PBAP onto mineral" should be "PBAP **on** mineral"

DONE

57. Line 507: Isn't is more likely the discrepancy between the MPOA-only simulation and the observations is due to the observations containing (mostly) measurements that are not MPOA? It would be informative to separate these single-INP type comparisons (Fig. 6a-c) and compare to similar observations. See Major Comment #4.

We followed the reviewer's suggestion, see our reply to major comment # 4.

58. Line 509: Wilson et al. (2015) did not use Chl-a to parameterize MPOA. SML INP concentrations were scaled by measured TOC, and this scaling factor was applied to modeled organic matter in sea spray.

We admit that these sentences are confusing, Chl-a parameterization is used in the model for the MPOA source calculation. As we have also restructured the manuscript and added an evaluation of the INP-relevant aerosols as well as a section 3.3. on uncertainties, the above discussion has been modified and now reads:

"The discrepancy between observed INP and INP_{MPOA} can be partially attributed to missing INP sources, especially dust, and partially to the uncertainty in the INP_{MPOA} simulations resulting from both the MPOA simulations and the INP_{MPOA} parameterization."

59. Line 509-512: The overestimation of Wilson et al. (2015) seen in McCluskey, Ovadnevaite, et (2018) was specific to observations of marine aerosol. Why would you expect a MPOA-only parameterization to overpredict total INP, when dust is much more IN-active? Are the observations around -25 °C expected to be MPOA INPs?

This discussion has been modified in the restructured revised version of the manuscript (see reply above).

60. Line 515: I think you mean the McCluskey, Ovadnevaite, et (2018) parameterization here, not Wilson et al. (2015).

done

61. Line 517-520: This sentence requires more explanation or clarification. Why does sea spray surface area require detailed organic composition information? What do you mean by "SS size variability that occurred due to marine biological processes"? SS will change size once in the atmosphere, but the only direct link to marine biology after emission would be through condensation of emitted gases. What "process is not parameterized in McCluskey et al. (2018)" that is needed? Since you do not implement or otherwise discuss the McCluskey, Ovadnevaite, et (2018) parameterization, this sentence can probably be removed.

This part of the discussion has been removed.

62. Line 523-527: See Raman et al. (2023) for a more nuanced discussion of why equating SML and SS INPs may be a poor assumption. Organic enrichment factors between the SML and SS are hugely variable based on the specific organic composition, in addition to the aerosol production mechanism. The evaluation of TM4-ECPL OC in Myriokefalitakis et al. (2010) indicated a general underestimation of POC, and virtually no seasonality in the model. Even if the Wilson et al. (2015) relationship (which included only Arctic measurements) between SML TOC and atmospheric INPs is globally applicable to SS, you have not evaluated any other parameterization(s), and so have not provided any direct evidence your model is "more realistic". And indeed, the studies which have evaluated both the Wilson (2015) and McCluskey, Ovadnevaite, et (2018) parameterizations generally find the latter works better. An overactive Wilson (2015) parameterization may be counteracting the underestimation of OC by the model. The more complicated approach may be theoretically more realistic, but only if all the component parameterizations are very accurate. Can you comment on this at the fairly coarse model resolution used here?

We thank the reviewer for this comment. Indeed, as shown in Myriokefalitakis et al. (2010) and in new Figure 2 and discussed in section 3.1.2, the model underestimates OC in the marine environment. Here we used the Wilson et al (2015) parameterisation to derive the INP_{MPOA} which overestimates the ice activation of MPOA and indeed this seems to partially balance the underestimate of MPOA. This is now added in the manuscript in section 3.3: 'McCluskey et al. (2018) have reported that the Wilson et al. (2015) parameterization for MPOA ice activity may overpredict INP_{MPOA}. Our model tends to slightly underpredict INP in the temperature range around -25°C, consistent with the earlier discussed underprediction of MPOA levels by the model. Thus, the overly active parameterization of Wilson et al. (2015), shown by McCluskey et al. (2018), may be counteracting the model's underestimation of MPOA.'

63. Line 533: Could this discrepancy also be attributed to comparing with observations that are not dominated by PBAP INPs?

Yes, it is possible that the observed total INP is influenced by different types of INP than those simulated. In addition, the deviation of simulated individual INP types from observations, shown in the new Figure S6 as a function of binned temperatures, indicates that at temperatures of about -25 to -30°C, both INP_D and INP_{PBAP} separately overestimate the observed INP. For warmer temperatures, Figure S6 clearly shows that a major contribution to the model overestimation of the observed total INP is due to INP_{PBAP}.

64. Line 536: Replace "ice efficiency" with "ice nucleating efficiency"

done

65. Line 537-539: A similar limitation is true of the Wilson (2015) parameterization, which uses data from a small number of Arctic SML samples and then generates a global parameterization using simulated (not observed) atmospheric INP concentrations. Some caveats or discussion is warranted in the preceding paragraph or in the methods, as was given here for the Tobo et al. (2013) parameterization for PBAP.

The following text has been added in section 3.3. on uncertainties: "Parameterisations and model evaluation are based on observations geographically and temporally limited and used for global estimates; therefore, their global applicability remains an open issue."

66. Line 544: "highest correlation is found" should be replaced by "highest correlation between simulated and observed INP concentrations is found".

done

67. Line 549: Add "of" between "correlation coefficient" and "0.88". Remove the reference to Fig. 6c, the correct Fig. 6d is referenced earlier in the sentence. Add "in addition to dust" in between "MPOA" and "improves".

done

68. Line 550-552: The sentence beginning with "Model results are more consistent…" simply repeats what was just said and is unnecessary.

The sentence has been removed

69. Line 554: "contributor" should be plural, "contributor**s**"

done

70. Line 573: Can you expand on what you mean by "model performance"?

This sentence in the uncertainties section 3.3 is rephrased: "The effects of atmospheric ageing on ice-nucleating activity are probably a minor source of error in this study, in comparison to the introduced bias from the employed INP parameterizations and model performance in simulated INP-relevant particles"

The evaluation of simulated INP-relevant particles is now a full section 3.1.2.

71. Line 580: add ", but not INP_{PBAP}" after "are included".

done

72. Line 591-594: Did you show a relationship between PBAP and "regional weather patterns" somewhere? Or one between MPOA and "regions of high sea spray and phytoplankton activity"?

No, we did not show such correlations but the parameterizations used for the sources depend on meteorological factors for PBAP and on sea-spray emissions and chlorophyll-a for MPOA. The sentences have been rephrased for accuracy.

"Simulated concentrations of INP from terrestrial bioaerosol vary with season and meteorological factors that affect both the source of PBAP and its ability to act as INP. ….. Simulated INP from marine bioaerosol is found primarily over oceans and coastal areas and dominates between 40°-90°S (Southern Ocean), with high concentrations in regions of high sea spray and phytoplankton activity that influence the source of MPOA."

73. Line 599: remove "at" between "mainly" and "low altitudes".

done

74. Line 600-601: This paragraph would flow better if you remove the first sentence beginning with "Therefore, we propose…" and combine the rest of the paragraph with the very short one in lines 595-599.

done

75. Line 602: replace "could" with "can likely", since you do not show any evidence for this, it is your speculation.

done

76. Line 611: Does "sulfate acids" mean "sulfuric acid"?

corrected

Figure Notes:

1. Are all the data in the figures (except for Fig. 6) from 2015? What is the annual variability between the concentrations and percentages shown, if so?

All data presented are multi-year results. In the new Figure 8 we now show the coefficient of variation (Figure 8b) calculated as the standard deviation of the annual mean columns to the multiyear annual mean and is expressed in percent as an indicator of this variability, which locally can exceed 150%, maximizing in the tropical and polar regions.

2. Consider removing Fig. 1, since you barely mention it in the discussion. And/or, add some explanation to the main text to describe the different INP concentration metrics shown in the upper right

The figure is now discussed in the introduction

3. Please clarify which INP concentration metric you are plotting in every figure. See Major Comment #2.

done

4. I'm not sure Fig. 4 adds much to the discussion. Could you instead do the seasonal pie charts currently in Fig. 4 for Fig. 3? Between Fig. 2 and 3, the main conclusions from Fig. 4 have already been discussed.

Seasonal pies and Figure 4 have been removed. Figure 3 has been replaced by the new figure 9.

5. Separating the pie charts in Fig. 3 and 4 into only NH and SH are a bit oversimplified if you want to discuss dominant INP sources/types. As you mentioned, the main sources of PBAP are equatorial. The Southern Ocean and Arctic are very small sources of INPs overall, but also have very different clouds (height, temperatures, SLW) than in the tropics and mid-latitudes. Perhaps having 3 categories, one from 60 °S to 60 °N, plus the Arctic (>60 °N) and Southern Ocean/Antarctic (<60 °S) would make the most sense in terms of INP sources and also cloud regimes.

New figure 8 is drawn following these recommendations.

6. Caption for Fig. 6:
    1. Line 1075: add "simulated" between "accounting for" and "mineral dust"
    2. Line 1076-1078: the dashed lines are hard to see, perhaps replace with "shaded region" in the text, since you also shade the Pt_1 and $Pt_{1.5}$ regions.
    3. $Pt_{1.5}$ appears to be missing from the legends
    4. Line 1079: the error bars are the uncertainty on the observed values and not the simulated ones? What kind of uncertainty measurement is plotted (standard error, 90% confidence interval, etc)?
    5. Line 1080: The 1 and 1.5 in Pt_1 and $Pt_{1.5}$ should be subscripted.
    6. Line 1081: Do the Pt_1 and $Pt_{1.5}$ values consider the error bar on the simulated value, or just the monthly mean when considering whether they agree with the observations?

Figure 10 caption (earlier Figure 6) now reads: Comparison of INP concentrations calculated at the temperature of the measurements against observations accounting for simulated mineral dust (a), MPOA (b), PBAP (c), mineral dust and MPOA (d), PBAP and mineral dust (e), and mineral dust, MPOA and PBAP (f). The dark grey dashed lines represent one order of magnitude difference between modelled and observed concentrations, and the light-grey dashed lines depict 1.5 orders of magnitude. The simulated values correspond to monthly mean concentrations, and the error bars correspond to the

standard deviation of the observed monthly mean INP values. The color bars show the corresponding instrument temperature of the measurement in Celsius (a-f). Pt_1 and $Pt_{1.5}$ are the percentages of data points reproduced by the model within an order of magnitude and 1.5 orders of magnitude, respectively. R is the correlation coefficient, which is calculated with the logarithm of the values. Diamonds correspond to measurements (Bigg, 1990, 1973; Yin et al., 2012) that are compared with the climatological monthly mean simulations. Circles indicate comparisons between temporally and spatially co-located observations and model results.

7. Figure S1 caption: Did you mean to reference Fig. 6 instead of Fig. 4?

8. The colorbar scale on Fig. S3 may be a little too large. It's difficult to tell that dust is significantly more prevalent than PBAP.

We thank the reviewer for the careful reading and the comments. Old figures have been redrawn for better quality and colors, new figures have been added to address the reviewer's comments and suitable for the restructured discussion of the manuscript.