REPLIES TO REVIEWER #1 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.

Review of "Assessing the global contribution of marine, terrestrial bioaerosols, and desert dust to ice-nucleating particle concentrations" by Chatziparaschos and co-authors in ACPD.

Summary:

In this study the authors use a global chemical-transport model driven by reanalysis meteorology to simulate the global distribution of ice nucleating particles (INPs) over a period of 6/7 years. The authors build upon the previous study of Chatziparaschos et al. (2023) who considered INPs from quartz- and K-feldspar-containing mineral dust particles. In this study the authors include two new INP types (collectively referred to as bioaerosols): primary marine organic aerosol (PMOA) associated with sea spray aerosol, and primary biological aerosol particles (PBAPs). The PBAPs considered in this study include terrestrial bacteria and fungal spores. Though INP sources of PBAPs can also include pollen, the authors choose to not represent this class due to uncertainties with its treatment, and a likelihood of low concentrations when compared to the other classes. The authors have put effort into sufficiently representing the emissions of the bioaerosols and their subsequent evolution in the atmosphere. Methods for representing the ice-nucleating ability of each INP species are based on existing parameterizations from literature. The model simulation is run from 2009 to 2016 to obtain temporally and spatially collocated INP concentrations for comparison with a global database of INP measurements. The authors compare the global (and seasonal) distributions of simulated INP from the three INP sources, highlighting locations and seasons in which each displays particular importance or sensitivity. Dust is particularly important in the northern hemisphere, especially at higher altitudes, and PMOA is important in the lower atmosphere of the southern hemisphere towards the polar region. This is consistent with other studies. The INP sourced from PBAPs shows particular importance at warmer freezing temperatures (when compared to dust) and across much of the lower latitudes where there is considerable (seasonally dependent) productivity from vegetation. Finally, the authors compare the simulated INP distributions with a global dataset of observations. The authors report that the model is most representative when dust and PMOA are considered the only sources of INP, whereas the addition of PBAPs to either dust or dust+PMOA acts to worsen the comparison. Reasons for this are discussed, including the potential for an over-estimation in the sensitivity of PBAPs to act as INPs. The authors conclude from this that although dust and PMOA produces the best representation for INPs in the climate system (as a whole), the inclusion of PBAPs is still recommended given their likely strong spatial and seasonal variability. I thoroughly enjoyed reading this and believe it will be a valuable addition for the community, but feel it needs restructuring into a more logical and meaningful sequence. I also believe a small amount of extra analysis in the simulated-vs-observed comparisons could be beneficial in drawing out the impact of PBAPs at warmer temperatures (which is where they display the most importance). I therefore recommend minor revisions before being accepted for publication. I have provided more details 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:

I have three primary areas of concern.

#1. To trust the simulated INP concentrations the reader must believe that the model is able to capture the precursor aerosol species, which in this case includes dust, sea spray / PMOA, and PBAPs. The evaluation of dust and PMOA is referred to as having occurred, but the reader is redirected to previous studies. There is no evaluation of PBAP concentrations, and no discussion as to whether the simulated concentrations are appropriate. There may be no observations to compare to, and in this case should be stated as such. I strongly recommend that the authors include a sentence or two for dust and PMOA that provides a quantitative summary of what the evaluations show. What is the RMSE for the comparison to observations? Does it display any particular bias? Did the evaluation show the model is representative of the distribution and seasonal cycle? I also recommend the authors include an evaluation (or reasons for its omission) of the simulated PBAP concentrations/distribution.

We did not present evaluation of the INP-relevant aerosol species because these were done in earlier publications (Vignati et al., Atmos Environ. 2009; Myriokefalitakis et al., Adv. Meteorol., 2010, 1–16, doi.org/10.1155/2010/939171, 2010; Biogeosciences,13(24),6519-6543, doi.org/10.5194/bg-13-6519-2016; Chatziparaschos et al., Atmos. Chem. Phys., 23, 1785–1801, doi.org/10.5194/acp-23-1785-2023, 2023). However, following the reviewer's suggestion, a new section 3.1 on the simulated distributions of INP-relevant aerosols (3.1.1) and their evaluation (3.1.2) is added in the revised version of the manuscript. This section contains 3 new figures:

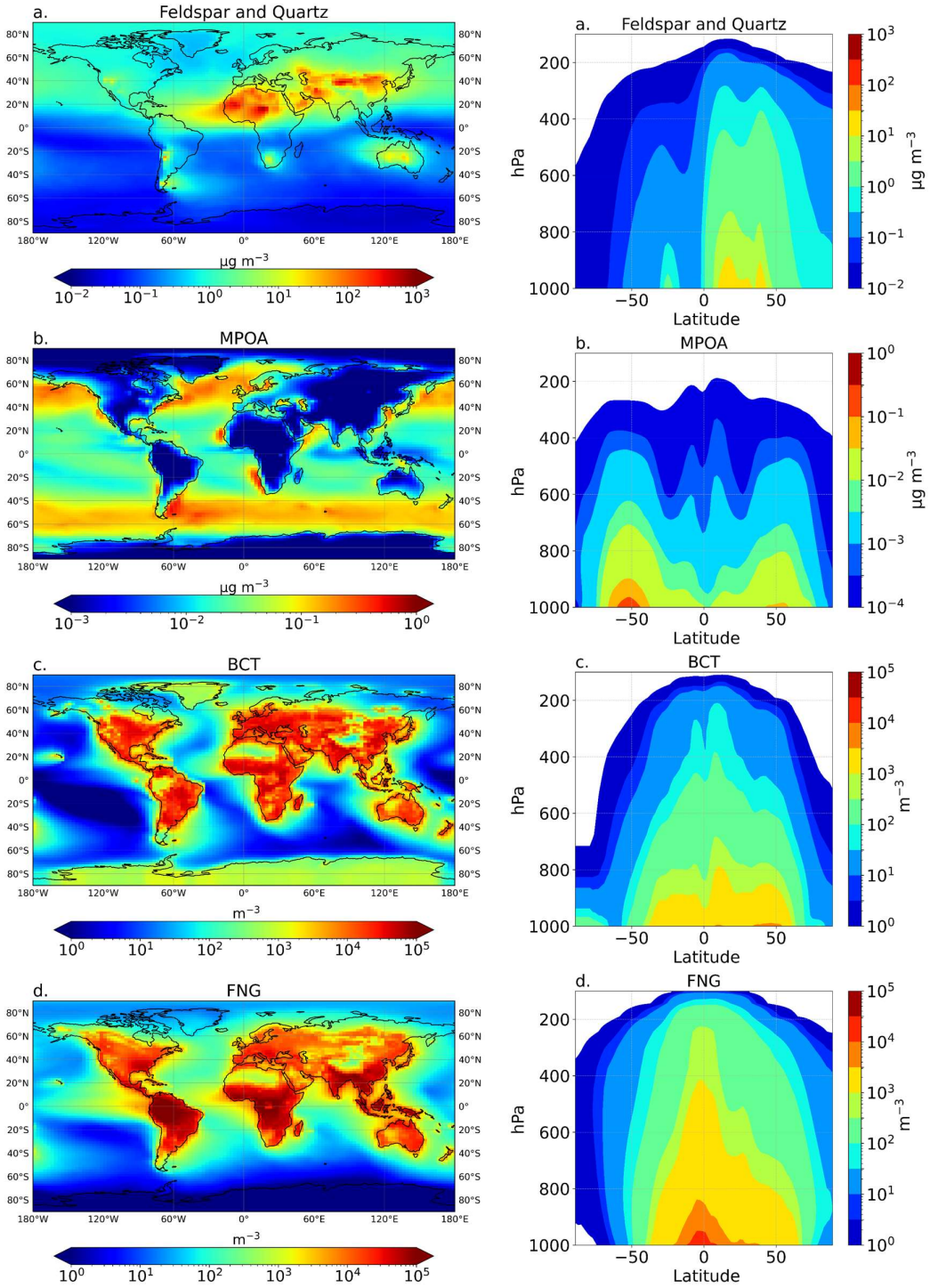

Figure *2*: Simulated multi-annual surface mean (left column) and zonal mean (right column) of (a) mineral dust (sum of feldspar and quartz) mass concentrations (μg m^{-3}), (b) primary marine organic aerosol mass concentrations (μg m^{-3}), (c) bacteria number concentrations (m^{-3}), (d) fungal spore number concentrations (m^{-3})

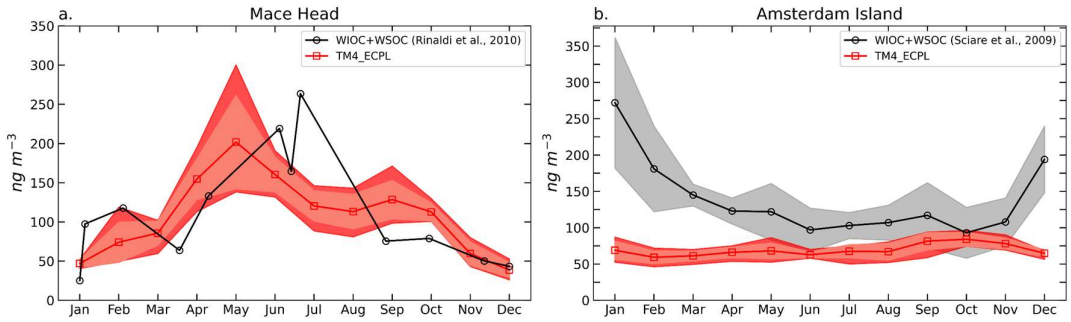

Figure 3: Monthly averaged concentrations of MPOA in ng-C m^{-3} at (a) Mace Head, (b) Amsterdam Island in ng-C m^{-3}. Observations from Rinaldi et al. (2010) and Sciare et al. (2009), respectively.

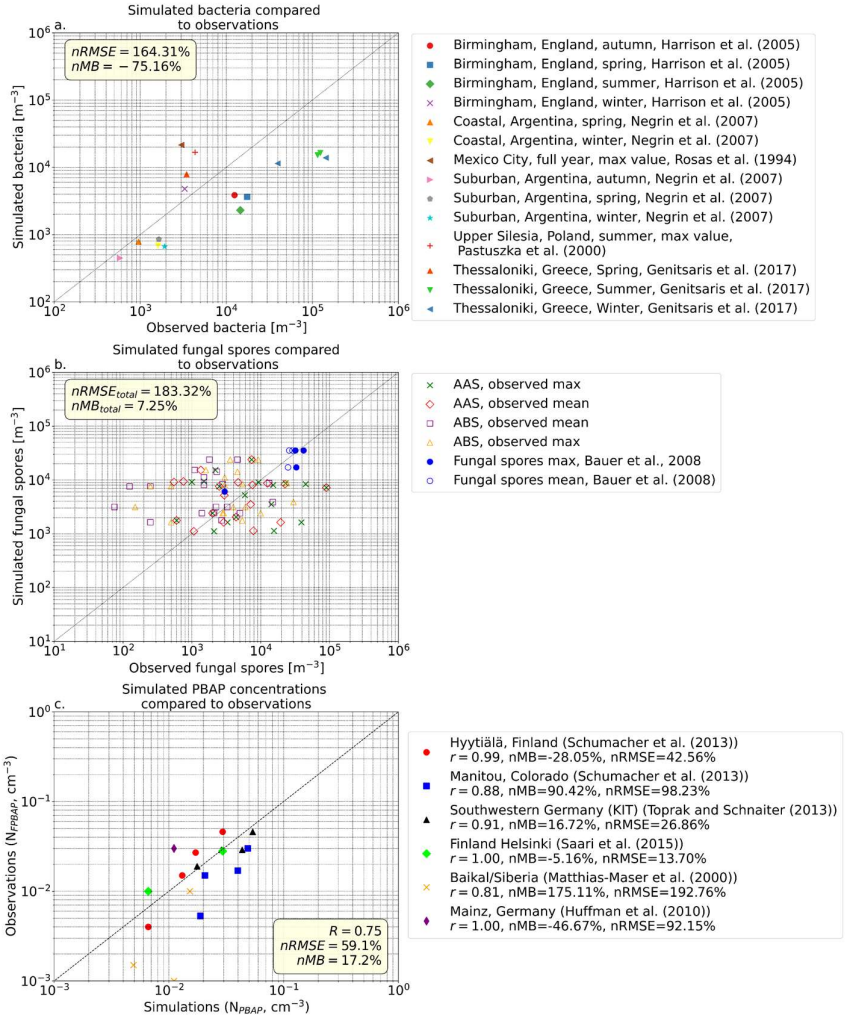

Figure 4: Simulated bioaerosol concentrations compared to observations. The observations are averages over different time periods, and the simulated data are taken for the corresponding months at the grid boxes containing the measurement site. (a). Bacteria concentrations; observations include concentrations listed in the supplement of Burrows et al. (2009) and Genitsaris et al. (2017). (b) Fungal spore concentrations, actively wet discharged ascospores (AAS) and basidiospores (ABS) compared to the simulated FNG. In the model, AAS and ABS are assumed to contribute one-third each to total fungal

spores. The observations are from the references listed in Elbert et al (2007) and Bauer et al. (2008). (c) Observations of fluorescent PBAP (FBAP) compared with model simulations of PBAP (for this comparison calculated as the sum of BCT, FNG and pollen). Observations are from the compilation of long-term measurements of atmospheric fluorescent bioaerosol number concentrations listed in Petersson Sjögren et al. (2023).

As well as two in the supplementary material:

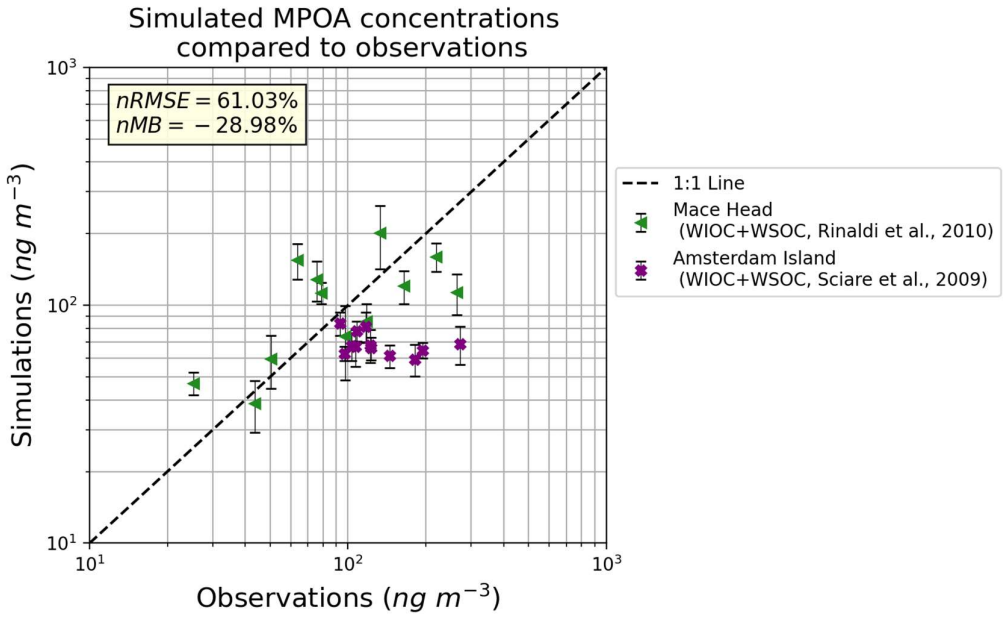

Figure S2: Simulated concentrations of MPOA compared with Mace Head (green) and Amsterdam Island (violet) measurements.

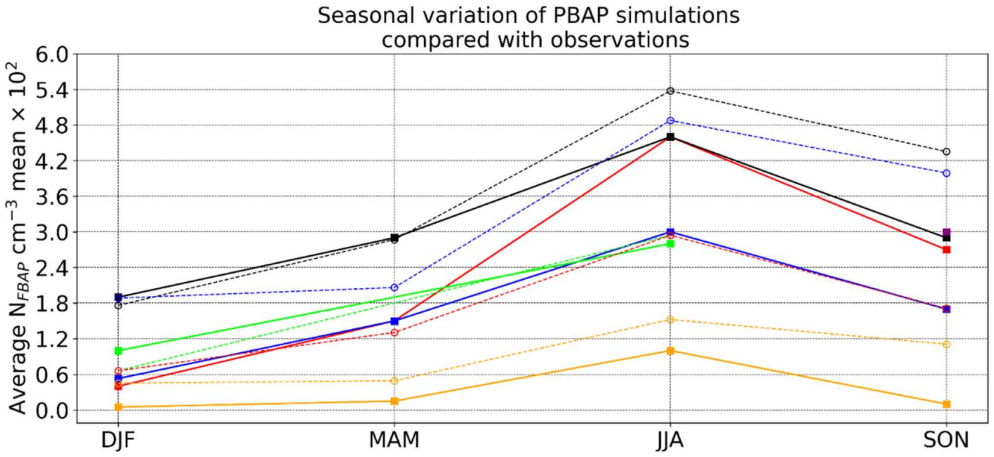

Figure S3: Seasonal variation of simulated number concentrations of PBAP compared to long-term (>4 weeks) FBAP observations compiled by Petersson Sjögren et al. (2023). Continuous lines are observations and dashed lines simulated number concentrations.

This new Section 3.1 concludes that "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 bacteria being significantly underpredicted and fungal spores moderately overpredicted. The model captures the seasonal variation of observed PBAP, while simulated MPOA in the SH shows no significant variation across the seasons. These model features have to 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. "

#2. This is my primary concern. The current structure of the results section is: Figures 2 – 4 present the relative importance of each INP species; Figure 5 establishes the global distribution of INPs in the model; and Figure 6 validates the INP model. At the moment this is backwards. The logical order is:

1. Here is our simulated INP model in its complete form (Figure 5; simulated INP distribution presented)
2. This is how it compares to the observations (Figure 6; INP model evaluated / validated)
3. Now we dig deeper into the relative importance of each species (Figures 2 – 4)

In this order the reader can then appreciate from the outset that the inclusion of PBAPs does not, in fact, tend to improve the representation of the global dataset of measured INP concentrations. This then has implications for the relative importance of PBAPs as is concluded from Figures 2 – 4. If, as suggested by the INP model evaluation, PBAP concentrations are too high or too ice-active, then this may suggest the 'relative-importance' analysis is biased towards INPs from PBAPs. The authors do a fantastic job at explaining the possible reasons why PBAPs do not improve the evaluation and I do not think that this means the relative-importance analysis needs to be removed. I think it is still entirely valid, but it should be highlighted that there is an associated uncertainty. The restructuring also applies to the abstract and conclusions section. I agree with the author's conclusions but feel they should be re-ordered for clarity.

We have restructured the presentation of the results as suggested by the reviewer. Section 3 of the results now starts with the presentation of INP-relevant aerosol distributions (new Figures 2-4 see our reply to comment #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 relative 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.

#3. In-line with comment #2 above I believe the INP model evaluation (Figure 5) would benefit from additional analysis. It is clear that PBAPs are active at warmer temperatures than dust. Therefore, if the INP model evaluation of dust+PMOA+PBAPs is made in binned temperature regimes does this show

better performance (when compared to dust+PMOA only) in the warmer temperature regimes? If so, this would strengthen the hypothesis that PBAPs are indeed important at warm temperatures. Also, the INP measurement database doesn't show particularly great global coverage. Given that the strongest signal from PBAPs is likely in low latitudes / boreal regions during the summer months, could the authors subset the comparison to these particular regions and seasons? This would provide a good basis for testing the importance of PBAPs.

We thank the reviewer for this comment. We have performed the additional analysis as suggested. We have added a new figure 9 an associate discussion in section 3.2.2 that further illustrates the different components of INP depending on atmospheric temperature, Figure 9 presents the percent contributions of INP types across three temperature ranges: [-40°C, -30°C], [-30°C, -20°C], and [-20°C, -10°C]. That figure highlights the spatial variability of the contribution of the studied species to the total simulated INP as a function of temperature. 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, independent of the altitude.

We have also produced Figure S6 with the deviation of INP model results from INP observations per temperature bin that has been added in the supplement and supports the conclusion on the overestimate of INP by PBAP in or model for the warmer temperatures. This figure is now discussed in section 2.3.3.

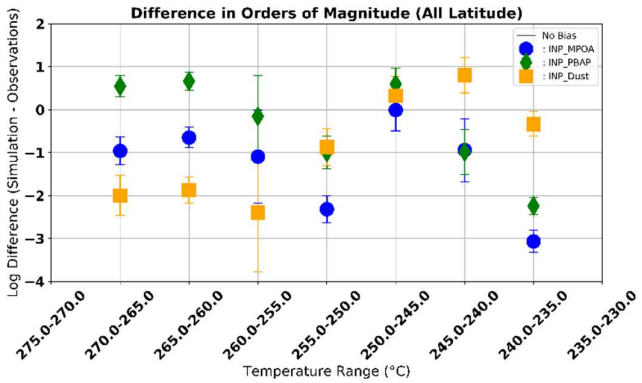

Figure S1: Order of magnitude / Logarithm of the deviation of simulated concentrations of INP types from total INP observations, as a function of binned temperatures. $INP_{\_Dust}$ in yellow, $INP_{\_PBAP}$ in green, $INP_{\_MPOA}$ in blue.

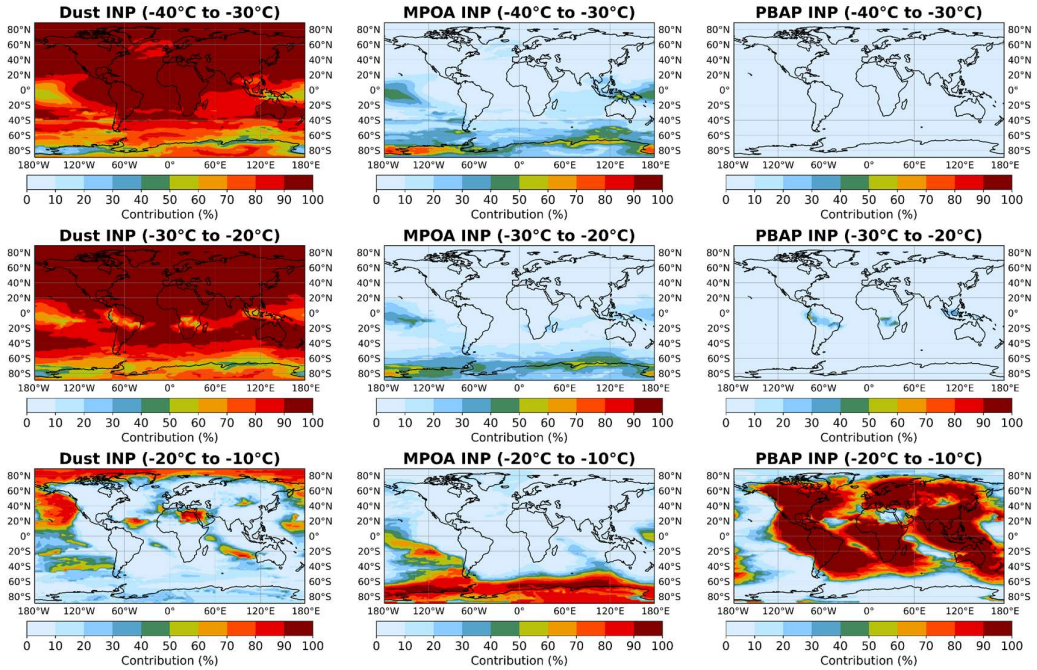

Figure 9: Multi-annual average percentage contributions of INP sources to the total INP column (surface up to 300 hPa): INP from mineral dust (left column), INP from MPOA (middle column), and INP from PBAP (right column), categorized by modelled temperature ranges: (top row) -40°C to -30°C, (middle row)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

We have also added a new Supplementary Figure S7 that shows simulated INP comparisons with observations per latitudinal zonal band and per INP type. This figure is discussed in section 2.3.2 and supports the regional importance of PBAP as INP.

"However, as seen in Figure S7, INP_{PBAP} compares well with INP observations in the NH and the extratropical SH (Figure S7 top and mid-rows). Therefore, in no case our results should diminish the role of INP derived from PBAP, especially in regions where PBAP is more abundant or ecosystems are diverse, such as the Amazon, affecting INP concentrations locally (Prenni et al., 2009). Most of the INP at warmer temperatures than -16°C are bioaerosols, suggesting that without improved representations of the sources and ice-nucleating activities of biological INP, models will struggle to simulate total INP concentrations at warmer temperatures and the resulting MPC"

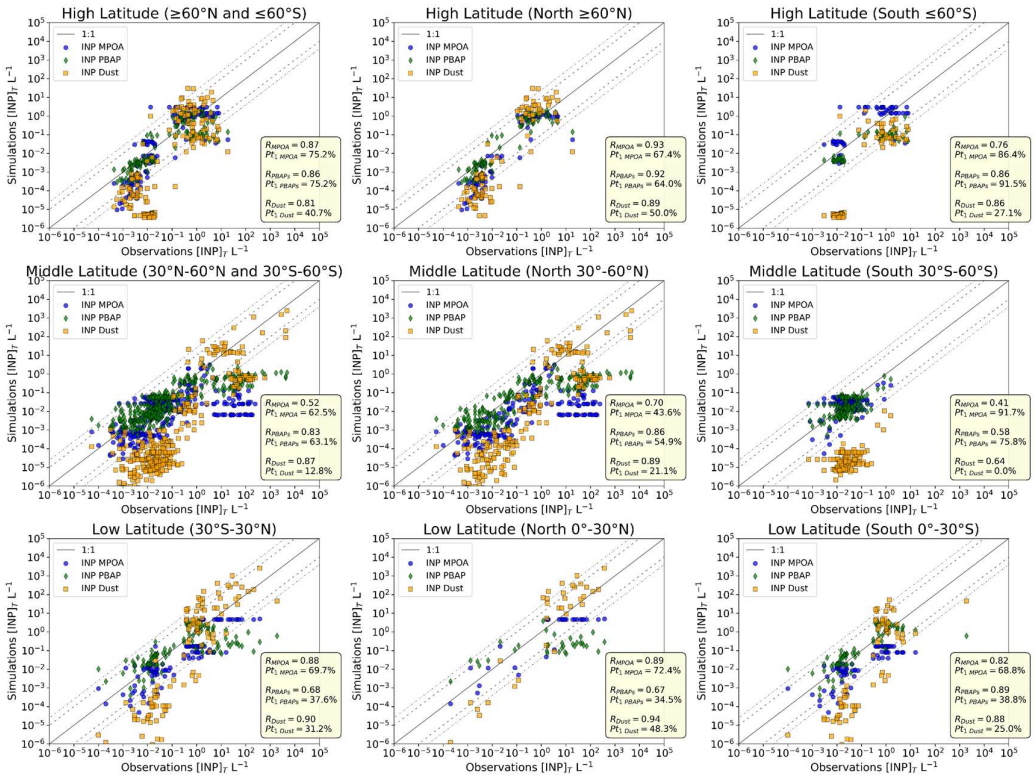

Figure S2: Comparison of INP concentrations calculated at the temperature of the measurements against observations accounting for mineral dust (orange), MPOA (blue), PBAP (green) separated in high, middle and low latitudes.

Minor comments:

Throughout: degree symbols are sometimes not superscript.

L30. As discussed in comment #2 the INP model evaluation should precede the more detailed analysis. Also, given the uncertainties I recommend changing “INP originating from PBAP…” to “The model suggests INP originating from PBAP…”

Modified as suggested

L50. MPC not yet defined in the manuscript.

It was done in the abstract, now also defined in the introduction

L54. “As a result, they significantly impact precipitation rates and radiative energy balance on both regional and global scales” Please include references to support this statement.

Hofer et al., Nature Communications Earth and Environment, 2024 https://doi.org/10.1038/s43247-024-01524-2 has been added

L56. Collection/collisional processes are also an ice-liquid interaction in MPCs. Can you expand to include other processes?

The following text has been added “In MPC, the most important ice formation process is immersion freezing, where an ice-nucleating particle (INP) becomes immersed in a supercooled droplet and initiates freezing, typically occurring between -5°C and -35°C (Pruppacher et al., 1998; Kanji et al., 2017 and references therein). Contact freezing happens when an INP collides with a supercooled droplet, triggering freezing on contact, often in turbulent conditions. This process is defined as separate from immersion freezing because of empirical evidence that some INP are more effective in this mode than when immersed in liquid (Shaw et al., 2005). Condensation freezing occurs when water vapor condenses and freezes simultaneously upon contact with an INP under specific supersaturation conditions. This process is less frequent compared to immersion freezing in atmospheric clouds (Vali et al., 2015). Deposition nucleation, where water vapor directly deposits as ice onto an INP without passing through the liquid phase, is more relevant at much colder temperatures (below -20°C) and is less significant in the typical temperature range of mixed-phase clouds (Kanji et al., 2017 and references therein) but it might still be important for cirrus clouds (Cziczo et al., 2013)"

Cziczo, D. J., Froyd, K. D., Hoose, C., Jensen, E. J., Diao, M., Zondlo, M. A., Smith, J. B., Twohy, C. H., and Murphy, D. M.: Clarifying the Dominant Sources and Mechanisms of Cirrus Cloud Formation, Science (80-.)., 340, 1320–1324, https://doi.org/10.1126/science.1234145, 2013.

Kanji, Z. A., Ladino Moreno, L. A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D. J., & Krämer, M. (2017). Overview of Ice Nucleating Particles. *Atmospheric Chemistry and Physics, 17*, 12177–12223. https://doi.org/10.5194/acp-17-12177-2017

Pruppacher, H. R., Klett, J. D., and Wang, P. K.: Microphysics of Clouds and Precipitation, Aerosol Sci. Technol., 28, 381–382, https://doi.org/10.1080/02786829808965531, 1998.

Vali, G., DeMott, P. J., Möhler, O., and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmos. Chem. Phys., 15, 10263–10270, https://doi.org/10.5194/acp-15-10263-2015, 2015.

L71. “..via the WBF process and multiply via SIP… affecting precipitation ranges, cloud properties, and albedo”. Please include references to support his statement.

‘precipitation’ has been removed and the statement is supported by the existing references (Georgakaki et al., 2022; Korolev and Leisner, 2020).

L80. Could you expand to describe the ‘seeder-feeder’ mechanism?

We extend that sentence to explain briefly the seeder-feeder mechanism : Text added “ where ice crystals from an upper cloud (seeder) fall into a lower-lying liquid cloud (feeder) and grow there by riming or vapor deposition via the WBF process”

L85. “In the absence of a well-established theory for heterogeneous ice nucleation…”. What about classical nucleation theory?

‘At temperatures warner than -35°C’ is added. The sentence now reads” In the absence of a well-established theory for heterogeneous ice nucleation at temperatures warmer than -35°C, INP prediction typically relies on empirical parameterizations that are subject to considerable uncertainty and challenges.

L127. “…with INP concentrations estimated at approximately 1 to 2 per litre”. Would be useful to have some context - how does this compare to dust?

This sentence now reads: 'Only a small fraction of biological material can trigger ice nucleation (Huang et al., 2021), with INP concentrations estimated at approximately 1 to 2 per liter (Pöschl et al., 2010). These values are, however, about 2 order of magnitude lower than the highest observed INP number concentrations from dust (Murray et al., 2012).

L134. SIP yet to be defined.

Done (secondary ice particles)

L137. "..warm parts of the mid-latitude clouds..". I do not understand what this is referring to. Please can you rewrite for clarity?

This has been rephrased and now reads: ' PBAP contribute to droplet freezing rates of clouds in the warmer parts of mid-latitude atmosphere'

L137. "..with about 1x10^-5 % of the global average ice nucleation rates.." this is also a confusing statement. Please can you rewrite for clarity?

The entire sentence has been rephrased as follows: Global simulations including biological particles have estimated that PBAP contributes to droplet freezing rates of clouds in the warmer parts of mid-latitude atmosphere, accounting for 1×10^{-5} % of the global average ice nucleation rates, with an upper-most estimate of 0.6% (Hoose et al., 2010).

L139. "..altitudes between 400 and 600 hPa..". Please use consistent quantities/units.

The sentence has been rephrased to : "Spracklen and Heald (2014) suggest that PBAP can dominate immersion freezing rates at altitudes corresponding to pressures between 400 and 600 hPa,"

L181. "bioaerosols". This term hasn't been defined yet.

This is now defined in the beginning of the 7th paragraph of the introduction which reads: 'While it is well established that dust particles are the most abundant source of INP at temperatures below -20ºC, it is equally well established that bioaerosols, i.e. atmospheric particles of biological origin, such as fungal spores, bacteria, pollen, and plant debris, are highly efficient ice nucleators at warm temperatures (Huang et al., 2021; Cornwell et al., 2022) and may have a substantial effect on ice crystal formation (Prenni et al., 2009).'

L191. The model is very coarse, both horizontally and vertically. Do the authors think this will have implications for the emission and dispersion of aerosols and modelled INP concentrations?

Indeed, the model has a relatively coarse resolution which enables to perform multiple multiyear global simulations on the existing computer facilities. The spatial resolution of the model implies that the simulated INP concentrations are more representative of a climatology of the background atmosphere than of the INP- related aerosol source regions. This is the reason we have performed multiyear simulations and we analyze the average of them. We have added the following sentence: "Therefore, when performing multiyear simulations, the model provides a climatological view of the troposphere."

L205. Please expand this sentence to briefly say what drives the dust emission sensitivity – i.e., wind speeds, surface moisture, etc

This sentence now reads as follows: "Desert dust emissions are calculated online as described in Tegen et al. (2002) and implemented as in Van Noije et al. (2014) accounting for particle size distribution and

based on vegetation type and cover, dust source areas, snow cover, soil moisture, and surface wind speed."

L215. As per comment #1 please expand and provide a short quantitative summary of the evaluation.

Following one of the major comments of the reviewer, the evaluation of the INP-relevant aerosols is now part of a new section 3.1.2. See our reply to major comment #1

L245. "FNG are assumed to be … 3um diameter with 1000 kgm-3 density". Is this appropriate? Do the authors have a sense of the uncertainty of this and its variability across the globe?

Indeed, this number has a large uncertainty but it is commonly used for fungal spores modeling. We consider that the sentence as was written points this uncertainty : "Bioaerosol sizes range from fine to coarse, but since their shapes are not accurately known, for the present work, FNG are assumed to be monodisperse spherical particles of 3 μm diameter with 1000 kg m^{-3} density (Hummel et al., 2015)." So, no change has been done.

Did the authors evaluate the simulated bacteria + fungal spore concentrations against observations? Is this a key observational dataset we are missing?

This is now included in the revised manuscript in the new Section 3.1.2, see our reply to major comment #1

L278. As per comment #1 please expand and provide a short quantitative summary of the evaluation.

Evaluation is now in an entirely new section 3.1. It first summarizes finding from published evaluations of dust and of MPOA and presents the evaluation of MPOA and PBAP simulations. (see reply to comment #1)

L298. Please include some statistics that summarise the evaluation. This evaluation is actually shown in Figure 6a so could be referenced.

Statistics on this evaluation together with discussion are provided in section 3.2.2 where the INP simulations are evaluated for each INP type separately and combined.

"Considering dust minerals as the sole INP types leads to underestimation against observations (Fig. 10a; Pt_1 about 24% and R=0.84). Even if dust is the most abundant aerosol in the atmosphere, the simulated dust-derived INP cannot predict the observed INP, especially at high temperatures (>-15°C) and relatively low INP levels, since mineral dust particles likely become ice-active only at low temperatures (Chatziparaschos et al., 2023; Cornwell et al., 2023). Notably, the results show that there is some overestimation for colder temperatures, which may be partly related to the overestimation of dust in the model. Findings in Figure 10a are largely in agreement with the literature and have been thoroughly discussed in Chatziparaschos et al. (2023)."

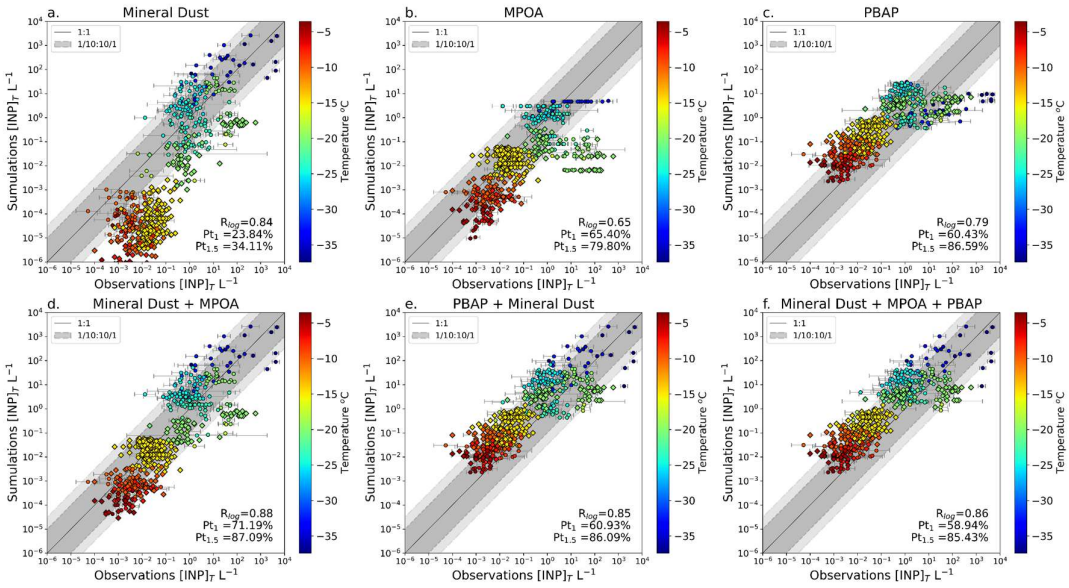

Figure 10: Comparison of INP concentrations calculated at the temperature of the measurements against observations accounting for simulated mineral dust (a), MPOA (b), PBAP (c), mineral dust and MPOA (d), PBAP and mineral dust (e), and mineral dust, MPOA and PBAP (f). The dark grey dashed lines represent 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 error 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.

L370. Missing minus sign.

corrected

L372. Specific to this line but more general. How relevant are the tropical and subtropical atmospheres for MPCs? Are they commonly observed here or more prevalent in the higher latitudes?

The following text has been added: "Note that MPC were found to occur more frequently in the convective towers in the tropics than at the mid-latitudes and the Arctic (Costa et al., 2017)."

Costa, A., Meyer, J., Afchine, A., Luebke, A., Günther, G., Dorsey, J. R., Gallagher, M. W., Ehrlich, A., Wendisch, M., Baumgardner, D., Wex, H., and Krämer, M.: Classification of Arctic, midlatitude and tropical clouds in the mixed-phase temperature regime, Atmos. Chem. Phys., 17, 12219–12238, https://doi.org/10.5194/acp-17-12219-2017, 2017.

L404. I suggest adding a value that signifies a threshold for "high pressure levels".

We have rephrased this part of the discussion because it is the temperature that affects the INP activity of the different types of INP. Now in section 3.2.2 we provide such information:

"INP_{PBAP} is the primary type of INP between −12°C and −20°C…. INP_{PBAP} contribution to the total INP is more than 80% between -12°C and -16°C. … At temperatures colder than -20°C, nonbiological

aerosol particles such as mineral dust are effective INP (Murray et al., 2012; Si et al., 2019) and dominate the INP population (Fig. 7a).”j

L497. “Even if dust is the most abundant… cannot predict the observed INP, especially at high temperatures”. Is this consistent with other studies?

Yes. In section 3.2.3 we now state: “Findings in Figure 10a are largely in agreement with the literature (e.g. Vergara-Temprado et al., 2018) and have been thoroughly discussed in Chatziparaschos et al. (2023). “

L502/3. “…largely in agreement with literature and have been thoroughly discussed in ..”. This shouldn’t be redirected to a different study. Please summarise the discussion from Chatziparaschos et al. 2023.

This paragraph now reads:

“Considering dust minerals Considering dust minerals as the sole INP types leads to underestimation against observations (Fig. 10a; Pt_1 about 24% and R=0.84). Even if dust is the most abundant aerosol in the atmosphere, the simulated dust-derived INP cannot predict the observed INP, especially at high temperatures (>-15°C) and relatively low INP levels, since mineral dust particles likely become ice-active only at low temperatures (Chatziparaschos et al., 2023; Cornwell et al., 2023). Notably, the results show that there is some overestimation for colder temperatures, which may be partly related to the overestimation of dust in the model. Findings in Figure 10a are largely in agreement with the literature (e.g. Vergara-Temprado et al., 2018) and our earlier study by Chatziparaschos et al. (2023). “

The evaluation of dust simulations by Chatziparaschos et al. (2023) is summarized in the new section 3.1.2.

L505. “… increasing the predictability of the model to 70%..”. Recommend including “increasing the predictability of the model FROM XX% FOR DUST to 70%...”

This sentence now reads: ‘INP_{MPOA} alone significantly improves the prediction of the observed INP at high temperatures, increasing the predictability of the model from 24% for dust alone to 77% for MPOA alone (Pt1, Figs. 10a and 10b).’

L506. I think this is where the additional analysis suggested in comment #3 would fit – and enhance the rigor of the evaluation.

Done – see our reply to major comment #3

L511/2. “The model tends to slightly overpredict INP only in the temperature range around -25C”. Could the parameterisation exhibit an incorrect temperature-dependence?

We have checked – the temperature dependences are correct.

L532. Have you tried looking at only-fungal-spore INPs vs only-bacteria INPs? Would this help identify which sub-species of bioaerosol is driving the discrepancy?

In the new section 3.1.2 we now evaluate separately bacteria and fungal spores. We conclude: “PBAP concentrations are also slightly overpredicted (by about 20%), with bacteria significantly underpredicted and fungal spores moderately overpredicted.” See figures in our reply to major comment #1

L600 onwards. In this paragraph the authors list a number of potential sources of uncertainty and reasons for model bias. What is required in order to constrain or address these? More laboratory measurements? More in-situ observations? More locations? Additional techniques to establish particle composition at sampling site? It would be good to identify what is currently lacking and is needed in future studies/campaigns.

Missing data availability statement.

We have added the following text at the end of the conclusion: "Due to the large variability of INP with space and time, effort must be put into acquiring more data on INP ambient levels as well as the ice nucleating properties of individual aerosol types as well as how these change with atmospheric ageing. These are needed to build regionally and globally representative datasets and reduce the uncertainty in the parameterizations of their sources in numerical models. Tackling these research priorities is essential for developing a more comprehensive understanding of the atmospheric variability of INP and their impacts in different cloud regimes and climate "

Figure 1. Reference to different methods of calculating INP concentration depending on convective state. This is not discussed in the manuscript, and therefore is quite confusing. Please include an explanation of this in the text. Also consider removing this figure as it does not add much to the manuscript and is only briefly referred to.

We provide Figure 1 as a summary of ice nuclei formation in the atmosphere. Therefore, we kept it and added appropriate explanation in the text: "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."

Figure 2. Viewing this figure feels like an eye test. Please increase the font size of all elements. Also, this would greatly benefit from distinct colormaps for the two rows. One for concentrations, the other for percentage contributions. The same comments apply to Figures S2 and S3 in the supporting information.

All figures are redrawn