

***Interactive comment on* “The contribution of fungal spores and bacteria to regional and global aerosol number and ice nucleation immersion freezing rates” by D. V. Spracklen and C. L. Heald**

Anonymous Referee #2

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In this work the authors implement an emission parameterization of primary biological aerosol particles (PBAP) into an aerosol transport model and use it to study the global distribution of fungal spores and bacteria. They also analyze their effect on Cloud Condensation Nuclei (CCN) concentrations and immersion freezing rates. The authors conclude that PBAP have a very limited effect on CCN concentrations and globally-averaged immersion freezing rates. However at lower altitudes PBAP may dominate immersion freezing rates.

The effect PBAP on cloud formation and climate is still surrounded with uncertainty and is often neglected in atmospheric models. However experimental evidence suggests

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that PBAP are ubiquitous and that their impact on climate is not negligible. The authors address an open question in the understanding of climate and their study is relevant to the atmospheric community. Unfortunately the method followed by the authors has several flaws (particularly regarding immersion freezing) that strongly affects their results and mislead their conclusions. The authors are also scant in the description of the modeling setup and the analysis of their results. I am afraid that in its present form this work cannot be recommended for publication in ACP.

1 General Comments

It is not clear why the authors decided to run their model at low resolution and only for a year. Clearly there is seasonality and interannual variability in vegetation and temperature that may affect the emissions of PBAP. The ECMWF reanalysis is available at much higher resolution and it is surprising that it was not used. The authors run a single year, however they make comparisons against long term observations which is clearly a flawed approach.

The authors could have done a much better job describing the implementation of the emissions parameterization. It is not clear how the Leaf Area index data from MODIS was used, whether the mapping is done every time step or changes during the year, and what is the temporal and spatial resolution of the data.

The analysis of the model results relies heavily on annual global means, which mask a lot of the variability and the importance of local effects. For example, the ecosystem dependency of the emissions parameterization will result in seasonal and interannual dependency of the spatial distribution of PBAP which is completely neglected. The importance of PBAP likely lies on being the only available CCN and ice nuclei in regions where neither soot nor dust are present. This is completely missed in a global mean calculation. The authors also make little discussion of the effect of the sensitivity of

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their assumptions in implementing the emission parameterization on their results.

The method used to determine immersion freezing rates is flawed. The authors completely neglect the fact that immersion freezing is a process exclusive of mixed-phase clouds (where ice and liquid coexist). Therefore the calculation of global immersion freezing rates without consideration of the presence of liquid water and whether the particles are incorporated in cloud droplets, is meaningless. This leads the authors to repeat the erroneous assessment of previous studies and conclude that PBAP do not have a significant effect on climate. Since most liquid water is found at temperatures where dust and soot are not active ice nuclei, it is likely that such conclusion is erroneous.

2 Specific Comments

Page 32460 Line 23. They can also be (and very likely are) important on a global scale.

Page 32462 Line 24. It should be mentioned here that recent studies (e.g., DeLeon-Rodriguez et al. 2013) suggest a significant contribution of bacteria to the particle population of the upper troposphere, with implications still unexplored.

Page 32463 Lines 1-2. Please spell out GLOMAP and TOMCAT.

Page 32463 Line 7. What is the temporal resolution of the ECMWF product used?

Page 32463 Line 7. This is fairly low resolution and must be justified since several important factors may be missing:

- How many levels does the model have between 700 hPa and 300 hPa, i.e, mixed phase conditions? More importantly, how many levels have mean temperature between 255 and 273 K, i.e., the conditions at which PBAP would have a greater effect. I suspect that the number may be quite low (about 3?), and wonder

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whether they may be enough levels to extract meaningful conclusions on freezing rates.

- Also, one will imagine that deep convection is an important mechanism of vertical transport of PBAP, but cannot be resolved at this resolution.
- A single year is quite a short time for a meaningful comparison against long term observations. Did the authors try a longer time period in any simulation? Why year 2000? Does interannual variability play a role in PBAP concentrations/freezing rates?

Page 32463 Line 23. What is the sensitivity of the results to the assumed sizes for bacteria?

Page 32464 Lines 2-6. These sentences are confusing and require more explanation. Is the MODIS-IGPB data a leaf area index climatology for the year 2000? If not, it is not clear what kind of data was used. Was the “mapping” done dynamically during the run, or was it done once and applied to all seasons?

Page 32464 Line 8. Are these values annual means?

Page 32464 Lines 18-20. Please provide references for such studies, and also the studies that show biases in the technique.

Page 32465 Lines 10-12. Please provide references supporting these statements.

Page 32466 Line 1. Does this mean that all fungal spores found in marine regions are advected from the land? Please clarify.

Page 32466 Lines 2-4. This is a confusing sentence. Please clarify.

Page 32466 Lines 16-17. This statement seems to indicate that the emitted particle size changes for different ecosystems. Please clarify.

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Page 32466 Lines 17. A sensitivity study showing results with a different size will help elucidate whether this is actually the cause of the overprediction.

Page 32467 Lines 1-8. There are several issues with this comparison. Please give more information about the observations, i.e., extend, coverage, type of ecosystem. Also, the model resolution is too low to make any meaningful comparisons against local observations. Finally, the model was run only for one year which hardly represents a seasonal climatology of PBAP emissions.

Page 32467 Lines 22-24. It is too superficial to just say that the patterns are similar to other studies. A deeper analysis will ask for example what is the origin of the observed patterns? What regions are likely to be impacted the most by PBAP emissions? What is the spatial distribution at different temperatures? Is there seasonal variability to such patterns?

Page 32467 Line 26. Seems that the assumed size of spores has a significant impact on the results. This should be explored in sensitivity studies.

Page 32468 Line 9. What properties are assumed for bacteria and fungal spores to calculate the CCN values?

Page 32468 Line 15. This depends on what conditions the authors are referring to. Please specify that this refers to mixed-phase clouds.

Page 32468 Lines 16-29. These calculation is misleading and the conclusions likely incorrect:

- Immersion freezing refers to the freezing of cloud droplets, and therefore depends on the droplet concentration. How is it possible to calculate immersion freezing rates for dust and soot at 100 hPa as shown in Fig. 4? If the likelihood of finding a water droplet is taken into account, then things will be very different, as PBAPs are likely to be more active ice nuclei at temperatures where droplets are more available. Thus they contribute a lot more to immersion freezing than what the

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Figure suggests.

- Part of the problem is the ice nucleation parameterization, which calculates immersion freezing rates regardless of the presence of droplets. The parameterization is also based on classical nucleation theory, which has issues, and the approximations made in its development are valid only at low freezing rates. Although it is out of the scope of this study to evaluate the parameterization, it is surprising that authors just take it for granted and did not explore the sensitivity of their results to the ice nucleation parameterization.
- Without accounting for the presence of droplets, the comparison of global immersion freezing is meaningless. Immersion freezing rates of dust at low temperature are of course dominant (if there would be droplets there) and bias the mean. A complete calculation would also account for the likelihood of finding a PBAP inside a droplet, which I suspect is much higher than for dust and soot.
- Since the modeling setup is limited (I don't think the ECMWF product provides droplet number concentration), I'll suggest that the authors find a way to limit their calculations to mixed-phase regimes (e.g., grid cells with liquid and ice present) and acknowledge that it still does not completely account for droplet number concentration, or do not discuss immersion freezing at all.

Page 32469 Line 16. Similarly as above, Figure 5 does not show the actual effect of PBAP on immersion freezing and leads to misleading conclusions. To correct it, the authors should limit their calculations to mixed phase regimes, weight immersion freezing rates by droplet number concentration, and account for the likelihood of finding a particle inside a droplet for each aerosol type.

Page 32469 Line 21. Figure 6 just follows the temperature distribution over the world. The Figure will be much more informative if the distribution is plotted at constant temperature (e.g. 255 K and 265 K) instead of at constant pressure.

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Page 32470. Many more things can be said about the distribution and effect of PBAP in the atmosphere. Discussion about seasonality, global variability, Ecosystem dependency and regions most prone to be affected by them is completely missing. Also I am confident that when the immersion freezing rates are corrected more meaningful conclusions regarding immersion freezing on PBAP can be derived from this study.

3 References

DeLeon-Rodriguez, N., Lathem, T. L., Rodriguez-R, L. M., Barazesh, J. M., Anderson, B. E., Beyersdorf, A. J., Ziemba, L. D., Bergin, M., Nenes, A., and Konstantinidis, K. T.: Microbiome of the upper troposphere: Species composition and prevalence, effects of tropical storms, and atmospheric implications, *P. Natl. Acad. Sci.*, 110, 2575–2580, doi: 10.1073/pnas.1212089110, 2013.

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