The contribution of fungal spores and bacteria to regional and global aerosol number and ice nucleation immersion freezing rates

Spracklen and Heald

Response to Review

We thank the editor for the opportunity to respond to reviewer comments and for taking the time to consider a revised manuscript. We also thank the reviewers for their time and for their comments on our manuscript. We respond to the reviewer comments in detail below. To guide the review process, reviewer comments are in italics, our responses are in normal text. We also attach a revised manuscript with tracked changes.

One reviewer suggests accept with minor revisions and the other accept with major revisions. We have been able to account for all the comments of the reviewers. We believe that our manuscript has been improved by this process and we hope that it may be acceptable for publication.

General comments

This manuscript by D. Spracklen and C. Heald picks up a study by myself and colleagues which was published in 2010 (Hoose et al., 2010), using a different model and adding analyses about supermicron particles, CCN and the regional distribution of immersion freezing rates. The main results of Hoose et al. (2010) are essentially reproduced. This is certainly reassuring, as it means that they are robust with respect to different treatments of the aerosol dynamics, the cloud microphysics, the dust and soot fields and the fungal spore emission function and size. The more subtle differences in the partitioning of the total freezing rate to the different species are appropriately discussed. The discussion of the regional contributions of PBAP to ice nucleation is essentially limited to one figure and one paragraph of the text. I would have liked to read more about this, especially as it is also emphasized in the title. The additional analyses of supermicron aerosol and CCN are also kept short but clear, maybe because the results were not thought to be very interesting, but I think this paper will be an important reference for these values which have never been calculated before.

The paper is well-written, very clear and a pleasure to read, and in my opinion it can be accepted for ACP once a couple of minor comments are adressed. Nevertheless, I think the article would be greatly enhanced if two points could be adressed which require some more work:

Quantification of the PBAP contribution to giant CCN

An analysis of the contribution to simulated freezing rates at the sites of the field campaigns which have identified significant numbers of bio-IN (e.g. Prenni et al.,

2009; Pratt et al., 2009; Prenni et al., 2013). This could be done e.g. as vertical profiles, seasonal cycles, ...
I leave it to the authors to decide whether or not these additional analyses are included.

We agree that quantification of PBAP to giant CCN and additional evaluation against field campaign observations are very important areas for future work. However, both these aspects would require substantial amounts of additional work. We therefore decide not to include these in our current manuscript and leave for future work.

Detailed comments

page 32460, line 23-25: I would like to point out that in Hoose et al. (2010), we stated: "However, these results do not rule out the local, regional and seasonal importance of biological ice nuclei." Thus, I don't think that there is any contrast between the findings presented here and our earlier results.
Thank you for pointing this out. We modify the last sentence in our abstract from:
"This demonstrates that PBAP can be of regional importance for IN formation, in agreement with case study observations but in contrast to recent global model studies that have concluded PBAP are unimportant as ice nuclei."
To:
"This demonstrates that PBAP can be of regional importance for IN formation, in agreement with case study observations."

page 32461, line 22 and elsewhere: When citing papers about fluorescent biological particles (FBAP), it should be carefully distinguished between FBAP and PBAP.
Changed as suggested. Supermicron number fraction of PBAP from Poschl et al. (2010) is 80% (FBAP fraction is 40%).

Page 32462, line 15: Please add that these results refer to one flight.
Added as suggested.

Page 32463, line 22: I strongly encourage the authors to publish the fit coefficients of their fungal spore emission function (Heald and Spracklen, 2009), such that they are available to the community.

We used emissions of fungal spores from the empirically optimised scheme of Heald and Spracklen (2009), where fine (E1) and coarse (E2) emissions are driven by leaf area index (LAI) and atmospheric water vapour concentrations in the surface layer (H2O_vap, v/v):
$E1=A1 \times LAI \times H_2O\_vap$
$E2= A2 \times LAI \times H_2O\_vap$

where, E in [g m^{-2} s^{-1}], $A1 = 5.17\times10^{-8}$ and $A2 = 1.55\times10^{-7}$.

Page 32463, line 27: Can you explain why you are going for the upper estimate?

Even with the upper estimate of emissions our model is typically biased low against available observations. We therefore choose the upper estimate as this gives us the closest match against observations.

The immersion freezing parameterization could also be mentioned in the "Methods" section instead of in the "Results" part.

We move the paragraph describing the immersion freezing methodology from the Results to the Methods.

Page 32466, line 23: I find the agreement and in particular the underprediction surprising as Burrows et al. (2009a)'s upper estimates for the emissions are used. Any comments?

The concentration of supermicron particles is sensitive to uncertainties in dry and wet deposition rates. We have added a sentence on the evaluation of supermicron particles in GLOMAP (see response to comment below), demonstrating that these are reasonably simulated by the model. However, there are also large uncertainties in emissions of dust and sea spray. So our evaluation does not preclude the possibility that supermicron removal mechanisms are too fast in our model. Unfortunately, insufficient data are available to test this, especially with regard to PBAP. What is more important for our study is that the simulated atmospheric concentrations of PBAP broadly reproduce available observations.

Page 32477: I assume that the observations in panel a) are from (Sesartic and Dallafior, 2011), please add.
Added.

Page 32467, line 8: How well are other supermicron particles simulated? Is there any reference in which they are evaluated for GLOMAP?
We add the following:
"Previous model evaluations have demonstrated GLOMAP reasonably simulates the mass and number concentrations of dust (Manktelow et al., 2010) and sea spray (Mann et al., 2012), giving us confidence in the distribution of other supermicron particle sources"

Page 32468, line 9: I would be curious which fraction of the bacteria and the small/large fungal spores are activated at 0.2% supersaturation.

We were remiss not to describe our method for calculating CCN. We add the following to the methods:
"CCN concentrations were calculated using the simulated aerosol size distribution and the approach of Petters and Kreidenweis (2007). We assign hygroscopicity parameters for sulphate (0.61, assuming ammonium sulfate), sea salt (1.28), black carbon (0.0), and particulate organic matter (0.1)."

Using this method, a supersaturation of 0.2% activates particles larger than about 80-150 nm (depending on composition). All bacteria and fungal spore particles will be activated as CCN at 0.2% supersaturation.

Could you also give numbers for the contribution to CCN at a lower supersaturation (i.e. giant CCN)?
This information is not provided by current model diagnostics, and would require repeat simulations to ascertain. While we agree that this is an interesting related question, it is not central to our study and would require considerable effort/computational time to address.

The simulated global mass burdens of bacteria and fungal spores would also be of interest for comparison to published values.
We add:
"We compare our calculated global annual mean mass burden of fungal spores and bacteria to that previously reported using the same PBAP emission schemes. The simulated global annual mean burden of fungal spores calculated here (0.15 Tg) matches that previously reported using GEOS-chem (0.18 Tg) (Heald and Spracklen, 2009) and CAM-Oslo (0.094 Tg) (Hoose et al., 2010b). The simulated global annual mean burden of bacteria calculated here (0.011 Tg) is also similar to previously reported by Burrows et al. (2009b) (0.0087 Tg) and simulated using CAM-Oslo (0.0043 Tg) (Hoose et al., 2010b)."

Are the immersion freezing rates shown here in-cloud values (if so, how is cloud presence diagnosed? RH? presence of liquid water?), all-sky values or are they calculated as a function of temperature only, irrespective of RH?

Immersion freezing rates are all-sky values. We clarify this in the text. We also calculate immersion freezing rates weighted by ice-cloud fraction (see response to Referee #2).

If the immersion freezing rates as shown here depend on the presence of clouds, liquid water or RH, then the cloud scheme is also relevant. I assume that the cloud are not influenced by the calculated immersion freezing rates. This could lead to some inconsistencies (e.g. high freezing rates in the lower parts of a cloud but plenty of water above that). This should be mentioned.

We previously presented all-sky values. We clarify this throughout the text. We use a chemical transport model, so meteorology is prescribed from reanalysis. We now also calculate immersion freezing rates weighted by ice-cloud fraction (see response to Referee #2).

Page 32469, line 25: This is interesting, but the question is how relevant these regions with very small immersion freezing rates are. The freezing rate of $10^{-14}cm^{-3}s^{-1}$ converts to less than $10^{-6}L^{-1}day^{-1}$, which is extremely small and probably irrelevant for cloud glaciation and precipitation formation. I understand that the tropics are not shown in Fig. 6 for exactly this reason, but how high are the total freezing rates in the regions which are plotted? I recommend to show results only for regions with a total freezing rate above a physically motivated lower limit. This also depends on the answer to how the averages are calculated.

We agree that different lower limits could be chosen. It is difficult to determine what lower limit would be irrelevant in the atmosphere. We therefore would prefer to retain our lower limit of 10^{-14} cm^{-3} s^{-1}. We add:
"We note that very small immersion freezing rates may have limited atmospheric impacts."

*I would add "regions where biological particles contribute substantially to **small** ice nucleation rates".*

Added as suggested.

Technical corrections
Page 32461, line 25 and page 32467, line 12: Matthais-Maser ->Matthias-Maser
Page 32464, line 25: Dallifor _> Dallafior

Done

Anonymous Referee #2

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 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 obervations which is clearly a flawed approach.

We use a standard spatial resolution for global chemical transport models and global atmospheric chemistry models.

It is true that there is likely to be interannual variability in the emissions of PBAP. However, variability in emissions is poorly understood and is not yet treated by PBAP emission schemes. For example, the bacteria emission scheme that we use is ecosystem dependent and does not calculate seasonal or interannual variability in emission. Exploration of interannual variability is therefore clearly beyond the current scope of available emission schemes. To acknowledge this we add the following sentence to our conclusions:
"We note that existing PBAP emission schemes have not been designed to adequately represent seasonal and interannual variability."

The observations we use are typically available for only one year so do not give information on interannual variability.

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.

We have clarified our implementation. The Burrows et al. emission scheme does not use LAI. The MODIS data is used to define ecosystem type (e.g., forest, crop, grassland etc). We add: "Note the emission scheme for bacteria does not include a dependence on LAI."

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 immersion freezing rates.

We respectfully disagree that we do not explore seasonal and spatial effects. We evaluate the seasonal cycle in fungal spore concentrations against available observations (Fig. 3). Insufficient data are available to sensibly evaluate the seasonal cycle for bacteria. We explore the spatial contribution of PBAP to immersion freezing rates during the NH summer (July).

The bacteria emission scheme parameterises emissions based on ecosystem type and does not have seasonal or interannual variability in emissions since the locations of ecosystems are fixed. Not enough is understood about interannual variability in bacteria of fungal spore

emission for us to explore this issue. Furthermore insufficient long-term data of PBAP concentrations are available to enable the model to be evaluated further in this context.

The effect PBAP on cloud formation and climate is still surrounded with uncertainty and is often neglected in atmospheric models. However experimental evidence suggests 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.
This is a good point. We thank the reviewer for raising this issue. We note that our method of calculating all-sky immersion freezing rates is analogous to the way that global mean CCN (i.e., potential CCN) are typically calculated in global modelling studies. We clarify throughout the text where our calculations are "all-sky". We also calculate immersion freezing rates weighted by ice-cloud fraction as suggested by the referee. See our detailed responses below. This slightly reduces the importance of PBAP at the global scale but does not alter our overall results or conclusion.

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

We have modified this sentence in response to comments from Reviewer #1. We agree that it is possible that PBAP has importance at the global scale. However, we do not think we have demonstrated this sufficiently in our paper to warrant a statement in the abstract.

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.
We add:
"PBAP can also make substantial contributions to supermicron particle number in the free and upper troposphere. DeLeon-Rodriguez et al. (2013) reported that bacteria can represent 20% of total particles in the 0.25 – 1 μm diameter range at ~10 km over the Atlantic Ocean."

Page 32463 Lines 1-2. Please spell out GLOMAP and TOMCAT.
We spell out GLOMAP. TOMCAT is no longer an acronym (see Chipperfield, 2006).

Page 32463 Line 7. What is the temporal resolution of the ECMWF product used?
We use 6-hourly files from ECMWF linearly interpolated onto the model time-step. We clarify this in the text.

Page 32463 Line 7. This is fairly low resolution and must be justified since several important factors may be missing:
This is standard resolution for global chemical transport models.

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 whether they may be enough levels to extract meaningful conclusions on freezing rates.
The model has 11 vertical layers between 700 hPa and 300 hPa.

Also, one will imagine that deep convection is an important mechanism of vertical transport of PBAP, but cannot be resolved at this resolution.
The model uses reanalysis meteorology from ECMWF which captures the large scale motion of the atmosphere. It is correct that individual deep convective events are not captured.

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?
We note that long-term (multiannual) observations of PBAP are lacking. We already comment that such observations would be useful (see Conclusions).

Interannual variability in PBAP emissions is not well understood. The bacteria emission scheme we use does not include any seasonal or interannual variability. Our fungal spore scheme (Heald and Spracklen, 2009) is based on LAI and specific humidity, so the scheme would produce interannual variability in emissions. However, long-term observations of fungal spores are not available to evaluate simulated variability. We therefore leave running longer simulations until we better understand such emission drivers and when multi-annual PBAP observations are available. For computational reasons we simulate a single year. We selected the year 2000 for no specific reason.

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

We apply the same emission size for bacteria as used in previous studies. Exploring the sensitivity to size would require additional simulations which are not possible at this time. We therefore leave this to future work.

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?

The referee misunderstands the emissions scheme, described in previously published work (Hoose et al., 2010a,b; Burrows et al., 2009a). The emission scheme is based on ecosystem type (e.g., forest, grassland) which does not vary seasonally. The MODIS IGBP data is a land cover classification used to define the spatial distribution of ecosystem type (see P32464, L5). The emissions scheme for bacteria does not depend on LAI. We clarify this in the text: "Note the emission scheme for bacteria does not include a dependence on LAI."

Page 32464 Line 8. Are these values annual means?
Yes, we clarify this.

Page 32464 Lines 18-20. Please provide references for such studies, and also the studies that show biases in the technique.
We add a reference to Burrows et al. (2009a).

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

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

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

Page 32466 Lines 16-17. This statement seems to indicate that the emitted particle size changes for different ecosystems. Please clarify.
We do not vary emitted particle size based on ecosystem type (see Methods). Our statement is merely suggesting possible reasons for model discrepancy. As stated, additional observations are required to explore this in more detail.

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.
As stated, additional observations are required to explore this issue further. We leave a detailed sensitivity study for future work.

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.
Details about the observations are given in Section 2.2).
Also, the model resolution is too low to make any meaningful comparisons against local observations.
See comments above about spatial resolution of the model.

Finally, the model was run only for one year which hardly represents a seasonal climatology of PBAP emissions.
See previous response.

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?
Our statement here confirms that our model simulates similar patterns to previous studies. We thank the reviewer for suggestions for future research.

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.
As stated above, we leave additional sensitivity studies for future work when additional observational constraints are available to validate.

Page 32468 Line 9. What properties are assumed for bacteria and fungal spores to calculate the CCN values?
We have now included details of how we calculate CCN concentrations:

"CCN concentrations were calculated using the simulated aerosol size distribution and the approach of Petters and Kreidenweis (2007). We assign hygroscopicity parameters for sulphate (0.61, assuming ammonium sulfate), sea salt (1.28), black carbon (0.0), and POM (0.1)."

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

Page 32468 Lines 16-29. These calculation is misleading and the conclusions likely incorrect:
The paramaterisations we apply are based on previously published work (Hoose et al., 2010a, b). We agree that there is substantial uncertainty in these calculations. We already acknowledge this uncertainty which we explore through a sensitivity study.

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 Figure suggests.
Our immersion freezing rate calculations are all-sky. We clarify this throughout the manuscript. We also report freezing rates weighted by ice-cloud fraction as suggested by the referee (see below).

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.
We agree with the referee that it is out of scope of this study to evaluate the parameterization. We carry out a sensitivity study to explore the importance of this uncertainty. A full uncertainty analysis would be useful and is left for future work.

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.

We note that our all-sky method of reporting freezing rates is analogous to the way that CCN are typically calculated in global modelling studies. Where we report all-sky numbers we clarify this. We also report freezing rates weighted by ice cloud fraction. We add the following text to the methods:

"We report immersion freezing rates in two ways: all sky and weighted by ice-cloud fraction. We apply monthly mean ice cloud fraction from the International Satellite Cloud Climatology Project (ISCCP) D2 data (Rossow and Schiffer, 1999) for the year 2000."

Calculating immersion freezing rates weighted by ice-cloud fraction slightly reduces the global contribution of PBAP to immersion freezing rates. We add the following:
"When we calculate immersion freezing rates weighted by ice-cloud fraction, global annual mean rates are still dominated by dust (97.2%) with smaller contributions from soot (2.8%), fungal spores (8.1×10^{-6}%) and bacteria (1.3×10^{-6}%)."

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.
Weighting by cloud fraction does not change this figure greatly. We prefer to retain Fig. 5 as all sky. We change Fig. 6 to be weighted by cloud fraction (see below).

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.
This is a good suggestion. We modify Fig. 6 to be for 260 K and 263 K, weighted by ice-cloud fraction. We modify the text to match the figure.

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.
Our study does not allow us to comment on many of the topics raised by the reviewer. We now calculate immersion freezing rates weighted by ice cloud fraction. This does not change the overall conclusions of our study. See responses to comments above.

References

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