

Response to Referee #1

Comment: This study explores the skill of three parameterizations for fungal spores, implemented in the EMAC model, to reproduce observed fungal spore counts, as well as (in combination with parameterized bacteria and pollen) fluorescence observations. Given the challenges in interpreting the observations (undercounting of spore counts, varying sensitivities of fluorescence), the study struggles to conclude as to the skill of these schemes as comparisons with the two datasets lead to opposite conclusions. Given this ambiguous result, I was disappointed that the authors did not pursue more exploration on the modeling side. I provide some specific suggestions below which would expand the utility of this study and ensure that it meets the standard for publication.

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestions, as described below.

1. Expand modeling

Comment: The results from none of the 3 fungal spore simulations is very satisfactory. Can the authors suggest (and possibly test?) improvements?

Response: Based on the spore counts data we collected, we could not find any significant correlation between observations and the usual meteorological parameters influencing the spores release such as specific humidity, relative humidity, temperature, etc..(see Jones and Harrison, 2003); therefore we couldn't build any statistical relationship based on these data. More long-term observations reporting spore counts (in the form of time series) are needed to be able to build a new emission parametrization.

Nevertheless, since the parametrization proposed by Hoose et al. (2010) has been scaled to match the Heald and Spracklen (2009) emissions, we recommend applying an additional scaling factor of 6, which is the median of the ratio HS concentrations to the spore counts. This has been added to section 3.1.

- **Comment:** Page 8,10: the authors claim that differences in bacteria from Burrows et al. may be the result of using the MODIS ecosystem distribution rather than the Olson distribution. This seems like something that could be easily confirmed with the model.

Response: Indeed this can easily be confirmed with the model but not shown here since we know that Burrows et al. (2009b) used exactly the same model EMAC and setup (all processes included here), the only difference between setups is the ecosystem distribution.

Comment: Page 13: the authors highlight the deficiency of not including seasonal or diurnal variability for bacteria. Could they perform a simple

sensitivity test to explore how imposing seasonal variation might impact their results?

Response: The bacteria emission parameterization developed by Burrows et al. (2009b), used in this study, does not include any information about diurnal and seasonal variation. This was due to a lack of such observations for different ecosystems.

A new emission parameterization for bacteria will be the subject of a future independent work that will include the newly available observational data published since the publication of Burrows et al. (2009a).

2. Aerosol size assumptions

Comment: Page 3, line 28: Why are fungal spores and bacteria treated as monodisperse? This seems an unrealistic assumption.

Response: Fungal spores, bacteria and pollen are treated as monodisperse for the sake of consistency with previous modeling studies using the same assumption (Burrows et al. 2009b, Hoose et al. 2010a and 2010b, Haga et al. 2014, Hummel et al., 2015, Twohy et al. 2016, Hummel et al. 2018). Since the size distribution does not affect the removal rate in the model, there is no need to speculate about it, and add potentially misleading information, as the latter is not available from measurements.

Comment: Page 3, lines 32033: comment on the size dependence (if any) of these processes

Response: the size dependence of these processes, has been described by Tost et al. (2006) and Kerckweg et al. (2006), and the sensitivity of atmospheric transport to particle size for the EMAC model has been tested and described in detail in Burrows et al., 2013 and Kunkel et al. (2013).

Comment: Page 5, line 17: 5 um seems large for fungal spores. Heald and Spracklen include fine and coarse mode particles, so this assumption does not seem consistent. Please comment.

Response: As mentioned earlier, we used the emission parametrization proposed by Hoose et al. (2010) adapted from the emission estimates of Heald and Spracklen (2009). This emission parametrization has been calculated based on the assumption of a mean spore diameter of 5um.

Comment: Page 8: While the authors claim that their results are not sensitive to the assumed size, it would certainly impact the conversion from number to mass. Might this help explain differences among previous fungal spore estimates discussed on page 9? If the authors feel these differences are not the result of assumed size, can they offer some explanation for these substantial differences? And why do the distribution and magnitude of bacteria agree better with previous studies than for fungal spores?

Response: We are not certain if the referee is referring to the comparisons with previous estimates of global total fungal spore emissions, at the bottom of page 8, or the comparisons of simulated fungal spore number concentrations with observed fungal spore number concentrations (spore counts), at the top of page 9.

On page 8 (l. 26-33), we compare with previous emission estimates that were reported on a mass basis in earlier studies. Compared with 17 Tg yr⁻¹ calculated in this study, Heald and Spracklen (2009) calculated 28 Tg yr⁻¹, and Hoose et al. (2010) calculated 31 Tg yr⁻¹, all when using the same emission parameterization. Hoose et al. (2010) used a mean spore diameter of 5 μm and Heald and Spracklen (2009) used the same diameter in the coarse mode.

These differences with previous fungal spore estimates are explained by the physical parameters in the emission scheme, i.e., Leaf area index and simulated surface humidity in the different host models.

By contrast, the different emission schemes formulate emissions differently, producing large discrepancies in simulated emissions between the different schemes.

On page 9, we discuss the comparison with number concentrations simulated when using these different emissions schemes, versus observed spore counts (number concentrations), shown in Figure 2. This comparison does not depend on the conversion of simulated number to simulated mass.

Comment: Figures 3a, 4a, 5a seem to suggest more export of pollen and bacteria than spores (though this may be a false impression due to the color bar). Can the authors confirm this by including global mean lifetime numbers for the 3 classes of PBAP? If lifetimes do differ, why is this the case when the authors indicated that removal processes are not dependent on size?

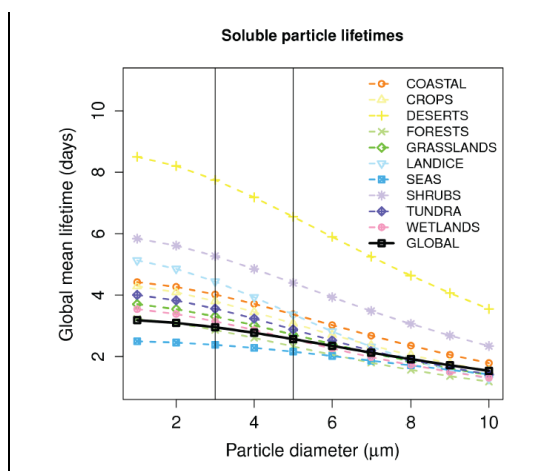
Response: We could say that, actually, the color bars are not easily comparable. The scales of fungi and bacteria must be multiplied by a factor of 1000, i.e. different from the scale of pollen. Thus, pollen is actually the aerosol category that is less transported. Based on the comments of Referee#2, we preferred to move the pollen figures to the SI.

Nevertheless, the reviewer's comments have helped us to realize that the dependence of transport and removal on particle size was not sufficiently explained in the original manuscript.

The export of particles simulated by the model varies as a function of both particle size and the geographic location of the emissions, which determines the atmospheric transport and removal processes the particles experience.

However, the differences in atmospheric residence time and export that are associated with particle size are on the order of a factor of ca. 1.5 – 2.5 when varying the particle size between 1 μm and 10 μm (Burrows et al., 2013, Figure

1, reproduced below for convenience). This is much smaller than the model-observation differences shown in Figure 2, which are frequently 2-3 orders of magnitude. A clarification has been added to the revised manuscript.



MINOR

Comment: 1. Page 1, Line 19: measurements are spores, not all PBAP

Response: This has been corrected.

Comment: 2. Page 1, Line 23: meaning of “reflects a greater difference” is unclear. Compared to what? Observations? Or do the authors mean the ratio of bacteria to fungal spores varies more widely? Please modify text.

Response: This part of the sentence has been removed for the sake of clarity.

Comment: 3. Page 1, lines 27-28: “of fungal spores and pollen”, why not include bacteria in this sentence?

Response: The contribution of the global bacteria mass concentrations to the total aerosol mass is too low (less than 1%) to be cited here.

Comment: 4. Page 6, line 9: what is the time span of the averaging? Is it possible to separate seasonal averages?

Response: Here the “averaging” means the averages over 4 years simulation. We provide a climatological value for each period of observation. All model data are sampled according to the time period of each observation.

Comment: 5. page 6, line 27: what is the upper size limit of these instruments?

The upper size limit of the UV-APS and WIBS is nominally 20 µm, though in practice the inlet design of an individual measurement site frequently lowers the upper size point somewhat.

Comment: 6. Page 10, lines 24-26: why are the Borneo and Nanjing data exceptional? The authors need to justify why they would remove these from the comparisons

Response: The Nanjing data reported too high concentrations that could be attributed partially to domestic pollution rather than biological particles (see the discussion related to these measurements). This has been added to the revised manuscript. For Borneo, we added it to the figure 6 with a different scale, and it has been included to the discussion.

Comment: 7. Section 3.5: the presentation of these results is a bit confusing. It would be helpful if the authors first discussed how many datasets are available for each season and commented on the observed seasonality before discussed the model performance.

Response: our point in this section is not to compare the seasonality of the model to observations. We believe that seasonality is not relevant here. The observations are only available for these specific time periods and our main objective is to compare the model concentrations to the available observations.

Comment: 8. Figure 6: It is very hard to see the data on this figure, suggest better use of scale (max value set to 0.1) to see data more clearly and using a color other than yellow/green which is hard to distinguish from white on the panels. The season labelling should be explained in the captions.

Response: We corrected the scale and changed the colors of the figure as the referee suggested.

Comment: 9. Page 11, line 1: unclear what “discrepancy” the authors are referring to. The measurements don’t distinguish these two classes of PBAP.

Response: indeed the measurements don’t distinguish between the two classes, but we expect from the measurements to see more fungal spores than bacteria (given all the uncertainties related to these measurements)

Comment: 10. Page 11, lines 25-28: clarify if these measurements are all for the same size ranges .

Response: Indeed, these measurements, as explained earlier in the paragraph and in Table 2, are for the same ranges.

Comment: 11. Page 12, line 19: reference of justification for assumed mass per particle needed

Response: The references have been added in the revised manuscript

Comment: 12. Page 12, line 20: specify that these means are for the surface

Response: The word “surface” has been added to the text.

Comment: 13. Page 12, line 28: The Poschl et al. numbers are averages over the deployment and only for supermicron particles and so cannot be directly compared to annual means of all aerosols from the model. Suggest that you compare to relevant month, coarse fraction only. If the large difference holds when the correct time of year is compared can the authors speculate as to why there would be such a substantial difference?

Response: We agree with the referee that this comparison is not appropriate therefore it has been removed.