

## 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.

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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

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**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.

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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.

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**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.

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**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?

5 **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).

10 2. Aerosol size assumptions

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

15 **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.

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

25 **Response:** the size dependence of these processes, has been described by Tost et al. (2006) and Kerkweg 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.

30 **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<sup>-1</sup> calculated in this study, Heald and Spracklen (2009) calculated 28 Tg yr<sup>-1</sup>, and Hoose et al. (2010) calculated 31 Tg yr<sup>-1</sup>, all when using the same emission parameterization. Hoose et al. (2010) used a mean spore diameter of 5µm 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.

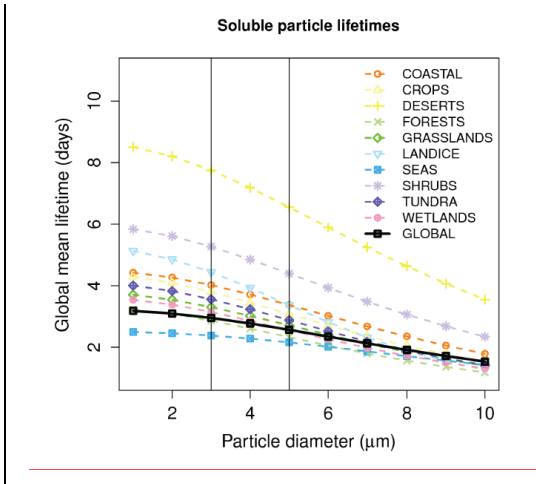
**Comment:** Figures 3a, 4a, 5a seem to suggest more export of pollen and bacteria than spores (though this may be a false impression due to the color bar). Can the authors confirm this by including global mean lifetime numbers for the 3 classes of PBAP? If lifetimes do differ, why is this the case when 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.

5 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.

10 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  $\mu\text{m}$  and 10  $\mu\text{m}$  (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

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

**Response:** This has been corrected.

20 **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.

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**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.

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**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 um, though in practice the inlet design of an individual measurement site frequently lowers the upper size point somewhat.

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**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.

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**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.

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**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.

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**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.

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**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 .

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**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

15 **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.

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**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?

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

## Response to Referee#2

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.

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### Summary

This manuscript has developed a global emissions and transport model for primary biological aerosol particles (PBAP). There have been several other prior attempts to do this for all types of PBAP (e.g., Jacobson and Street, 2009) as well as individual types (e.g., fungal spores Heald and Spracklen 2009; bacteria Burrows et al 2009a/b). In this regard, the work is necessary but the manuscript itself does not make new advances in our simulations or understanding of global PBAP. In general, the manuscript has several major omissions of the data and methods driving the model, and this makes it not possible to interpret the results in any meaningful way. This paper requires major revisions to be acceptable in ACP.

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### Major comments

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1. The manuscript states to have simulated the three main types of PBAP, yet there is no detail in the manuscript about the pollen emissions. The authors need to decide if are going to retain the pollen section of the PBAP inventory. If the authors choose to continue to include pollen in their total PBAP assessment, there are several new sections that are absolutely necessary to understand what how the authors are simulating emissions and when and where they might be important. These include the following:

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**Comment:** a. Add a section 2.2.3 for pollen. Section 2.2 is titled “PBAP emissions,” with one subsection devoted to bacteria (2.2.1) and the other to fungal spores (2.2.2), yet there is no corresponding section on the pollen emissions. A section explaining the pollen emissions parameterization is absolutely required. Similarly, this should include the size distribution implemented in the model (as is for fungal spores and bacteria).

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**Response:** We apologize for this omission in the text. The section has been added to the revised manuscript.

**Comment:** b. Provide some useful discussion on the pollen atmospheric distributions and their realism. For example, there is no discussion about the pollen emissions distributions that they simulate despite including a figure of pollen (Section 3.4). Specifically, their model simulates the highest concentrations of pollen in the tropics, which is inconsistent with the plant distribution of wind-driven pollen. Most plants in the tropics use insects or birds for transmission, so it is not expected that

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there would be high emissions in these locations. It is impossible to determine why this is without the explanation of the pollen emissions model (see point above).

**Response:** The emissions depend on LAI similar to fungal spores, therefore, it should not be unexpected that concentrations are high in the tropics. It is difficult to discuss the realism of the global pollen distribution, as an important effort is needed to collect a global database for pollen. The Jacobson and Streets (2009) provided the first pollen parametrization to be used for a global model. This parameterization has not been evaluated against observations for any global model, and has been mostly used by the community to provide a global distribution for pollen, despite of its many deficiencies (e.g. absence of the plant phenology and the land cover type). This was not the objective of the present study, as we focused on fungal spore distribution. In the discussion section, we recommend the use of a more recent parametrization developed and evaluated by Wozniak and Steiner (2017). For these reasons, we decided to remove Figures 5a and 5b from the main text and added them to the Supplementary Information, but we keep section 3.4, to be able to discuss the contribution of pollen to the total mass aerosol composition.

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**Comment:** c. There are ground-based pollen count observations of similar spatial sparsity to fungal spores. Why is a comparison of measured versus modeled not included?

**Response:** Our purpose in this study is to focus on fungal spores parametrizations and to less extent bacteria. Pollen has been added to model as an additional PBAP only in order to estimate their contribution to the total aerosol mass (see section 3.6). A further modeling study using the new pollen emission parameterization proposed by Wozniak and Steiner (2017) will be the object of a following study including comparison with the available pollen count observations, especially in Europe, where a large pollen counts database is available.

20 **Comment:** d. In the final discussion, there is hardly any recommendations or future work regarding these emissions and improvements. Again, if the authors choose to keep pollen in there, a more rigorous discussion is required to explain the role of this specific type of biological particle.

**Response:** Recommendations have been added to the discussion section.

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**Comment:** 2. There is no information about the LAI distribution used in the model. This is rather important for the fungal spore discussion, as much of the explanation for the three different fungal spores is often tied back to differences in leaf area.



**Response:** Both the HS and HU parametrization in our simulations use the same LAI distribution (as well as the pollen parametrization). Information about the LAI distribution used in our simulation has been added to the revised manuscript in section 2.1.

5 **Comment:** 3. Section 3.2: This type of correlation seems rather obvious: modeled LAI is of course going to be lower in urban areas, so then another factor would have to compensate (and it would be a meteorological parameter). I'm not sure how this would not be taken into account already by the existing models. If the authors think that this is important, then they should explore this in greater detail.

10 **Response:** We found these results interesting because they show the importance of the difference between urban and non-urban observations and their relationship with these physical parameters (usually not measured). This information could be taken into account potentially in future observations. Unfortunately, we could not find any observation publication reporting about these differences.

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**Comment:** 4. Section 3.5: For the FBAP comparisons, why not also include the pollen and see if that improves any observations? The caveat of why FBAP may not (page 6, lines 20-21) or may (page 10, lines 10-13) is rather confusing.

20 **Response:** As shown in figure 5a, the pollen number concentrations are much lower than bacteria and fungal spores concentrations, therefore their contribution to the total PBAP concentrations will be very low. Besides, as explained in Page 10, the upper size limit of the UV-APS and WIBS is nominally 20  $\mu\text{m}$ , though in practice the inlet design of an individual measurement site frequently lowers the upper size point somewhat, therefore pollen can not be included for comparison with FBAP observations.

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Minor comments

**Comment:** 1. The title should reflect the full acronym of PBAP (e.g., add the term "aerosol")

30 **Response:** The word has been added to the title.

**Comment:** 2. Abstract Lines 26-27: Needed?

**Response:** We believe that these lines summarize one of the main findings of this paper, therefore they should be kept in the abstract.

**Comment:** 3. Page 2 Line 19: fungal spores reference about being the most abundant and genetically diverse – this is a rather old reference, is there any more modern support for this idea?

**Response:** The reference to the work of Fröhlich-Nowoisky et al. (2009) has been added

**Comment:** 4. Page 2 Line 35: Global and regional models are cited here, yet the papers primarily refer to global studies on fungal spores and bacteria. There is a wealth of literature out there on pollen, and this should be included if pollen is kept in the manuscript.

**Response:** Actually, Jacobson and Streets (2009), whose pollen parametrization has been used here and Hoose et al. (2010) include both pollen modeling.

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**Comment:** 5. Page 5 line 31: Because the model was run without meteorological nudging, some brief reference to prior met evaluation of the model to indicate to readers that biases in the concentrations are not due to meteorological parameters such as temperature.

**Response:** References to the model evaluation has been added in section 2.1., as recommended by the referee.

**Comment:** 5. Page 4, section 2.2.1: More detail on the bacteria emissions, specifically the fact that they are constant and are not simulate with any meteorological dependency should be made very clear in this section. This is discussed later in the manuscript (e.g., Section 3.5) but it would be clearer to provide more detail here such that a reader is not looking up all the references.

**Response:** For the sake of clarity, we provide a table of the best-estimates fluxes in the Supplementary Material, and a reference to it in section 2.2.1.

**Comment:** 6. Page 4, line 29: what does “best estimate number fluxes” mean in this context?

**Response:** “best-estimate” is used considering the optimization method used by Burrows et al. (2009b) for the emission estimates to fit the observed number concentrations.

**Comment:** 7. Page 6, line 18: What are some examples of “highly fluorescent particles of non- biological origin,” and would these be more likely to be observed in anthropogenically influenced areas?

**Response:** This sentence “such as certain kinds of aged brown SOA, diesel soot particles and some HULIS types” has been  
5 added to the text.

**Comment:** 8. Section 3.1: Note several references to Figure 1 that should be Figure 2, also, the fit metrics are not displayed as stated in the text (lines 12-13).

10 **Response:** This has been corrected in the revised manuscript.

**Comment:** 9. Section 3.1: Please clarify what model layers are used to compare to observed fungal spore counts, as this also may affect the model evaluation.

15 **Response:** The word “surface” has been added to the mean number concentrations.

# Global modeling of primary biological aerosol particle concentrations with the EMAC chemistry-climate model

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**Abstract.** Primary biological aerosol particles (PBAPs) may impact human health and aerosol-climate interactions. The role of PBAPs in the earth system is associated with large uncertainties, related to source estimates and atmospheric transport. We used a chemistry-climate model to simulate PBAPs in the atmosphere including bacteria, fungal spores and pollen. Three fungal spore emission parameterizations have been evaluated against an updated set of spore counts synthesized from observations reported in the literature. The comparison indicates an optimal fit for the emission parameterization proposed by (Heald and Spracklen, 2009), although the model significantly over-predicts fungal spore concentrations in some locations. Additional evaluation was performed by comparing our combined bacteria and fungal spore simulations to a global dataset of fluorescent biological aerosol particle (FBAP) concentrations. The model predicts the sum total of measured PBAP concentrations relatively well, with an over- or under-prediction of less than a factor of 2 compared to FBAP. ~~The ratio of bacteria to fungal spores reflects a greater difference~~ However, the simulated bacteria concentrations outnumber the simulated fungal spore concentrations in almost all locations. Further, the modeled fungal spore results under-predict the FBAP concentrations, which are used here as a rough proxy for spores. Uncertainties related to technical aspects of the FBAP and direct-counting spore measurements challenge the ability to further refine quantitative comparison on this scale. We estimate that the global PBAPs mass concentration (apart from desert dust and sea salt aerosols), i.e. of fungal spores and pollen, amounts to 19% and 52% of the total aerosol mass, respectively.

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## 1 Introduction

Primary biological aerosol particles (PBAPs) are diverse and include bacteria, fungal spores, viruses, pollen as well as fragments of other organisms. Their size and source characteristics are different throughout the globe. This influences their residence times and spatial distributions. The increasing interest in PBAP is related to the effects they may have on agricultural crops, human health and atmospheric chemistry (Deguillaume et al., 2008). Airborne bioparticles may influence climate by acting as ice nuclei in mixed-phase clouds. The abundance of atmospheric ice nuclei (IN) can influence cloud development, impacting cloud radiative properties and the location and timing of rain formation (Bangert et al., 2012; DeMott et al., 2010; French et al., 2018; Prenni et al., 2007). PBAPs are likely to be important contributors to ice nuclei abundance under clean (or near-pristine) conditions in remote locations, for example over the Amazon rain forest (Pöschl et al., 2010; Prenni et al., 2009). PBAPs have been shown to make an important contribution to the atmospheric aerosol mass in some locations (Bauer et al., 2002; Després et al., 2012; Elbert et al., 2007). Since they widely range widely in size from approximately 0.01 to 100  $\mu\text{m}$ , the time suspended in the air can vary from minutes to days (Després et al., 2012; Fröhlich-Nowoisky et al., 2016; Pöschl, 2005). Microorganisms have also been shown to be transported over long distances with desert dust from Asia and Africa (Griffin et al., 2007; Prospero et al., 2005; Smith et al., 2013), potentially contributing to the global transport of genetic information.

PBAPs have been identified and characterized using a large range of methods, including traditional methods such as microscopic analysis and cultivation methods, and modern methods using molecular techniques (Caruana, 2011; Després et al., 2012; Griffiths and Decosemo, 1994). However, continuous measurements of PBAPs have been limited, and therefore actual abundances, properties, as well as the origin of PBAPs and their components are still poorly quantified and understood (Burrows et al., 2009a; Burrows et al., 2009b). Fungal spores are the most abundant and the most genetically diverse PBAPs in the atmosphere (Fröhlich-Nowoisky et al., 2009; Lacey, 1981). Fungal spores are also of critical importance because many species can induce considerable economic losses, acting as plant pathogens or triggering respiratory diseases and allergenic processes in humans (Reinmuth-Selzle et al., 2017). Inhalation of spores in significant quantities causes various respiratory diseases such as allergic rhinitis, asthma, and other allergic reactions (Burge and Rogers, 2000; Bush and Portnoy, 2001). More than 100 species have been shown to contribute to respiratory disorders (Green et al., 2005).

Estimates of the total global emissions of fungal spores emitted into the atmosphere diverge greatly across the literature, varying from 8  $\text{Tg yr}^{-1}$  (Sesartic and Dallafior, 2011) to 186  $\text{Tg yr}^{-1}$  (Jacobson and Streets, 2009). Fungal spores contribute up to  $\sim 45\%$  of the coarse particulate matter over the tropical rainforest (Després et al., 2012) and their number and mass concentrations are typically about  $10^3$  to  $10^4 \text{ m}^{-3}$  and  $\sim 1 \mu\text{m m}^{-3}$ , respectively (Fröhlich-Nowoisky et al., 2016). The number and composition of airborne fungal spores depends on complex interactions between biological and environmental factors, including the climate and local ecological systems (Grinn-Gofron and Bosiacka, 2015). Meteorological factors are known to influence their production, release and transport, which is contingent to geographical areas, vary seasonally, and interactions

with PBAP can also depend on the emitting species involved (Elbert et al., 2007; Fröhlich-Nowoisky et al., 2009; Hirst, 1953; Levetin and Dorsey, 2006; Li and Kendrick, 1995; Oliveira et al., 2009).

Global and regional models have been used to evaluate PBAP emissions, transport and their impact on the hydrological cycle by acting as CCN and IN (Burrows et al., 2009a; Heald and Spracklen, 2009; Hoose et al., 2010; Hummel et al., 2015; 5 Jacobson and Streets, 2009; Sesartic and Dallafior, 2011; Spracklen and Heald, 2014). These models require the credible representation of the emissions and particle properties influencing transport and removal from the atmosphere. Large uncertainties in the number concentrations remain, especially due to inherent uncertainties in the emission estimates of PBAPs.

The goal of this study is to evaluate three fungal spore emission parameterizations available in the literature and compare 10 their number concentrations, simulated with a global model, to an updated set of observations synthesized from the literature. We use a chemistry-climate model to simulate the total PBAPs present in the atmosphere including bacteria, fungal spores and pollen. We compare our simulated PBAP concentrations to a global dataset of fluorescent biological aerosol particles (FBAP) concentrations. These measurements have been performed with real-time techniques that detect the fluorescence signal through UV excitation of fluorophores commonly present biological materials (e.g., fungal spores, 15 bacteria, and leaf fragments).

## 2. Materials and methods

### 2.1. Model description

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The global ECHAM5/MESSy Atmospheric Chemistry – Climate (EMAC) model (ECHAM version 5.3.01, MESSy version 2.5.2 (Jöckel et al., 2005; Jöckel et al., 2016) was used to simulate the emissions and transport of biological particles. The primary biological aerosol particles have been included with the emission parameterizations described below.

Removal processes of particles simulated by the model include sedimentation, dry deposition, impaction scavenging, and 25 nucleation scavenging by liquid, mixed-phase, and ice clouds. PBAP dry and wet depositions are treated as described for other aerosol species (see (Burrows et al., 2009a; Pozzer et al., 2012; Pringle et al., 2010a) and references therein). We simulated the transport of aerosol tracers of different sizes, as described in more detail below. All particle classes are treated as having a lognormal distribution with modal-scale parameter  $\sigma = 1$  ( $\sigma = 1.4$  for pollen) and with a density of  $1 \text{ g cm}^{-3}$ . We assume that all particles can become activated as cloud condensation nuclei when calculating particle removal processes by 30 wet deposition, as described in (Burrows et al., 2009a). All PBAPs are transported as passive tracers, i.e., their concentrations are influenced by model processes such as dry deposition and scavenging by clouds and rain, but do not interact with radiation or change cloud microphysical properties. The sedimentation and dry deposition of the particles are treated as described in (Kerkweg et al., 2006). Wet deposition of the particles is described in (Tost et al., 2006). **We use the global Leaf Area Index (LAI) distribution by (Deng et al., 2006) to calculate the fungal spores and pollen emission fluxes.**

For the present study, we applied EMAC in the T63L31 resolution; with a spherical truncation of T63 (corresponding to a grid of approximately 1.9° x 1.9 ° in latitude and longitude, or approximately 140 x 210 km at middle latitudes), with 31 vertical hybrid pressure levels up to 10hPa. The model was run for five consecutive years without meteorological nudging from the year 2000 until 2004. AMIP-II monthly sea surface temperatures were used to provide boundary conditions for the atmospheric circulation, available for the period since satellite observations are available (1979). Climatological averages for PBAP distribution for the last four years of the simulation were used after a 1-year spin-up period. **The EMAC model, evaluated in (Jöckel et al., 2005; Jöckel et al., 2016), and used in similar configurations, has been shown to be capable of realistic simulations of aerosol transport and deposition for the transport of African dust to Europe (Glaser et al., 2012) and radioactive aerosol particles from the Chernobyl accident (Lelieveld et al., 2012).** We emphasize that the simulation results represent a climatology rather than specific weather conditions under which some PBAB samples may have been collected, hence we expect mean number concentrations and distributions to be represented by the model rather than distinct measurement data.

## 2.2. PBAP emissions

### 2.2.1. Bacteria

Bacterial emission fluxes are calculated using the **constant** best-estimate values from (Burrows et al., 2009a) for different ecosystems, which were optimized toward overall agreement with best-estimates of observation-based near-surface number concentrations (see Table SII). We used the MODIS International Global Biosphere Program (IGBP) global land cover classification to determine the spatial distribution of 18 different ecosystems. We lumped the categories defined in the MODIS classifications to match similar sets of lumped ecosystems used by (Burrows et al., 2009a) (i.e., derived from the Olson ecosystem types), with the exception of the “urban” ecosystem, which is only present in MODIS data. We used a geometric mean diameter for bacteria of 4 µm for continental sources (forests, shrubs, grasslands, wetlands, savannahs and urban ecosystems) and 1.4 µm for marine sources. These choices are based on values reported for the count median diameter of bacteria-carrying particles, which may include bacteria borne by larger particles such as dust and leaf litter and/or clumps of bacteria (Shaffer and Lighthart, 1997; Tong and Lighthart, 2000, 1999; Wang et al., 2007). **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 um and 10 um (Burrows et al., 2013). The sensitivity of atmospheric transport to particle size for the EMAC model has been tested and described in detail in (Burrows et al., 2013; Kunkel et al., 2012). Therefore we note that modeled transport and removal processes are not strongly dependent on the particle size, at least not in the lower**

$\mu\text{m}$  size range, so that we do not consider the simplified size attribution of PBAPs to be a limiting factor in the representation of atmospheric processes.

### 2.2.2. Fungal spores

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We compare three fungal spore emission parameterizations previously used in global and regional modeling studies. Firstly, fungal spore emission fluxes have been derived by (Heald and Spracklen, 2009) (HS hereafter) from an empirically optimized scheme where emissions are linear functions of the LAI (Leaf Area Index) and the specific humidity  $q$  at the surface. In order to match their emission estimates, (Hoose et al., 2010) applied the following formulation to calculate the emission flux in  $\text{m}^{-2} \text{s}^{-1}$ , assuming a mean spore diameter of  $5 \mu\text{m}$ :

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$$F_{H\&D} = 500 \text{m}^{-2} \text{s}^{-1} \times \frac{LAI}{5} \times \frac{q}{1.5 \times 10^{-2} \text{kg kg}^{-1}}$$

Note that this formulation has been scaled to match the emission estimates by HS for a mean spore diameter of  $5 \mu\text{m}$ .

The second parameterization we tested uses the emission number fluxes of fungal spores calculated by (Sesartic and Dallafior, 2011) (SD hereafter) for five different ecosystems (defined by (Olson et al., 2001)). We use the best-estimate number fluxes weighted by the area fraction of the respective MODIS ecosystems in the gridbox  $E_i$ . Note again that similar ecosystems from MODIS data are lumped according to the corresponding Olson ecosystems defined by (Sesartic and Dallafior, 2011). The total emission flux for fungal spores is given as in  $\text{m}^{-2} \text{s}^{-1}$ :

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$$F_{S\&D} = 194 \text{m}^{-2} \text{s}^{-1} \times E_{tropicalforest} + 214 \text{m}^{-2} \text{s}^{-1} \times E_{forest} + 1203 \text{m}^{-2} \text{s}^{-1} \times E_{shrub} + 165 \text{m}^{-2} \text{s}^{-1} \times E_{grassland} + 2509 \text{m}^{-2} \text{s}^{-1} \times E_{crop}$$

20 The third parameterization, derived by (Hummel et al., 2015) (HU hereafter), is adapted to measurements of airborne fluorescent biological particles across northern Europe. Similar to the parameterization of (Heald and Spracklen, 2009), this recent parameterization depends on LAI and specific humidity, and is extended to include temperature  $T$ :

$$F_{FBAP} = b_1 \times (T - 275.82) + b_2 \times q \times LAI$$

where  $F_{FBAP}$  the emission flux in  $\text{m}^{-2} \text{s}^{-1}$ ,  $b_1 = 20.426$  and  $b_2 = 3.93 \times 10^4$ ,  $T$  is the surface temperature in K,  $q$  the specific humidity in  $\text{kg kg}^{-1}$  and LAI the leaf area index in  $\text{m}^2 \text{m}^{-2}$



For each parameterization, the mean diameter was assigned according to the recommendation made for each in the three studies: 5  $\mu\text{m}$  for HS, 3  $\mu\text{m}$  for HU and 3  $\mu\text{m}$  for SD. We reiterate that these size classifications are not expected to significantly influence the results.

### 2.2.3. Pollen

- 5 For pollen emissions, we use the emission parameterization proposed by (Jacobson and Streets, 2009) for use in global models. We apply it in a simplified form given by (Hoose et al., 2010), neglecting dependence on time of the day, relative humidity and turbulent kinetic energy:

$$F_{\text{pollen}} = 0.5\text{m}^{-2}\text{s}^{-1} \times \text{LAI} \times R_{\text{month}}$$

- 10  $R_{\text{month}}$  is a factor accounting for seasonal variations (0.5 in the Northern Hemisphere for October to March, 2.0 for April to June, and 1.0 for July to September; and shifted by six months in the Southern Hemisphere). We assume a mean pollen diameter of 20  $\mu\text{m}$ . We stress the simplified nature of this parameterization. We have included it for consistency, and to obtain an approximation of the relative roles of different PBAP categories.

## 2.3. Data description

### 2.3.1. Spores counts

- 15 We compare the fungal spore concentrations calculated by EMAC using the three emission parameterizations described above, to observations collected through literature review. (Sesartic and Dallafior, 2011) have reviewed more than 150 studies and found that only a relatively small number, 35 of these, reported total fungal spore concentration measurements, excluding observations that employed cultivation of a subset of species (e.g., in a petri dish) and measurements that report only mass concentrations instead of spore counts. We updated this dataset with observations that meet the same criteria,
- 20 mostly from studies published since 2011. Our updated review revealed that much of the relevant literature reports only concentrations of the genetic diversity of fungal taxa and not their total concentrations, which explains the scarcity of data that can be used for model evaluation. The uncertainties related to these methods are discussed in detail in (Sesartic and Dallafior, 2011). The observations used for comparison with model results are listed in Table SI2. Modeled concentrations have been sampled from the output to match the period of observation for each location. Since we do not compute actual
- 25 meteorology but rather climatological conditions, our model results do not represent instantaneous local processes, especially when they vary strongly on a small scale. However, we expect that time averaging limits such biases. We differentiate the data by ecosystem using the MODIS categories and the description provided by the reference. Most of the observations that met our criteria for inclusion have been taken in urban areas.

### 2.3.2. FBAP observations

Over the last two decades real-time measurement techniques have provided opportunities to monitor airborne PBAP continuously at relatively high time-resolution. Techniques involving laser/light-induced fluorescence (LIF) have been particularly effective in rapidly providing information about PBAP in real time (e.g. (Fennelly et al., 2018; Huffman and Santarpia, 2017; Kaye et al., 2005; Pan et al., 2009; Saari et al., 2014; Sivaprakasam et al., 2009)). Among many available instruments, two commercially available LIF biosensors have been widely applied to ambient bioparticle monitoring and have helped to reveal fine detail about atmospheric PBAP patterns not previously observed (Gabey et al., 2010; Huffman et al., 2013; Huffman et al., 2010; Perring et al., 2015; Schumacher et al., 2013). For example, the ultraviolet aerodynamic particle sizer (UV-APS; TSI, Inc.) and the wideband integrated bioaerosol sensor (WIBS; University of Hertfordshire or Droplet Measurement Technologies) both characterize biological particles in real-time based on the intensity of fluorescence emission observed from individual particles after pulsed excitation at wavelengths characteristic for common biofluorophores (Foot et al., 2008; Hairston et al., 1997; Pöhlker et al., 2012). Despite the uncertainties related to this type of measurement (Huffman et al., 2012; Pöhlker et al., 2012; Savage et al., 2017), FBAP detected by the UV-APS or WIBS have been successfully used in some cases as a lower-limit for the atmospheric abundance of PBAPs in the super-micron ( $> 1 \mu\text{m}$ ) size range (Huffman et al., 2010).

In this context, however, it is important to mention a few caveats implicit with the assumption linking FBAP to PBAP. First, real-time LIF instruments can only detect the physical properties of particles (i.e., fluorescence and size) and cannot directly determine whether a particle is of biological origin. By applying certain analytical strategies, however, a given ensemble of particles may be assigned as PBAP with varying degrees of certainty. In some cases weakly fluorescing biological particles can escape LIF detection (e.g. (Huffman et al., 2012)) and in other cases highly fluorescent particles of non-biological origin, such as certain kinds of aged brown SOA, diesel soot particles and some HULIS types, e.g. can interfere with LIF detection to overestimate PBAP (e.g. (Gabey et al., 2013; Huffman et al., 2010; Saari et al., 2013; Savage et al., 2017)). Bioparticle size also plays an important role in LIF detection. For example, viruses are generally too small to be detected by LIF instruments, and almost all species of pollen are too large to be detected without fragmentation or instrument modification (O'Connor et al., 2011). Additionally, technical differences in instrument design, the choice of detection channels, and operational parameters can have significant effects on the reported number concentration of FBAPs and the quality of their correlation with PBAP classes (Savage et al., 2017). Nevertheless, we use the FBAP numbers reported by UV-APS and WIBS instruments as a rough proxy for PBAP, comparing the observed FBAP numbers both with more direct PBAP measurements and with model outputs.

We show FBAP observations using UV-APS and the FL3 channel from the WIBS-3 and WIBS-4A instruments (Table 2).

The majority of FBAP data shown were extracted from published reports without additional analysis or as tabulated by previous reviews (Fennelly et al., 2018; Saari et al., 2015; Yu et al., 2016). All original data sources are attributed in Table 2. By limiting WIBS data to only the FL3 data, the number concentration is expected to be significantly lower than total FBAP numbers often reported, but are used here for better correlation with the UV-APS due to similarities in fluorescence excitation and emission bands (Foot et al., 2008; Pöhlker et al., 2012; Savage et al., 2017). For all LIF data, mean FBAP number concentrations were integrated from either 0.8 or 1.0  $\mu\text{m}$  to 15 or 20  $\mu\text{m}$  and are reported for each of seventeen geographic locations (see Table 2 and Fig. 6) for a comparison with our simulated mean number concentrations of fungal spores and bacteria produced from the model discussed here.. The FBAP observations show a peak in the number distribution at about 1 – 4  $\mu\text{m}$ , irrespective of location or instrument. It has been previously suggested for several geographic locations that the UV-APS and WIBS FL3 channel may yield lower limit proxies for fungal spores, due in part to the large number concentration of spores in this size range compared to other biological particles (Gosselin et al., 2016; Healy et al., 2014).

### 3. Results

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#### 3.1. Comparison of the EMAC fungal spore concentrations with spore count observations

Our simulated, globally distributed, annual **surface** mean number concentrations using the three emission parameterizations are compared to the set of observations at various locations in Figure 2. Observational data are differentiated by biome as defined by MODIS data, including the urban ecosystem. The modeled concentrations are overestimated by all three parameterizations, but least by HS, which agrees better with observed spore counts, with a correlation coefficient (R) of 0.24 and a median model-to-observation ratio of 6, compared to more than 100 for the other two parameterizations. Two additional metrics of model-observation agreement are presented in Figure 2: the modified normalized mean bias (MNMB), a measure of bias that is symmetric with respect to over-estimates and under-estimates (ranging between -2 to 2, and equal to 0 for a “perfect” model), and the fractional gross error (FGE), a measure of relative model error, ranging from 0 for a “perfect model” to a maximum value of 2, which behaves symmetrically with respect to under- and overestimation, without over-emphasizing outliers (Huijnen and Eskes, 2012).

From the three model versions, HS performs best on all three scores. SD and HU show similar MNMB and FGE, but SD shows a slightly negative correlation with the observations. Although we compare local measurements limited in their representativeness in time and space to the relatively coarse grid size (approximately 140 km) of the climatological model data, only the comparison with HS is satisfactory. Surprisingly, the SD derived fungal concentrations are the least comparable to observations despite the fact that the formulation of the emission parameterization is based partly on these observations. This might be due to the change in the global ecosystems distribution, as we used the MODIS ecosystem,

which the HS parameterization was based on, instead of the Olson distribution, which the SD parameterization was based on. The HU emission parameterization might not be suited for use in global modeling studies since it has been optimized for a regional modeling study over northern Europe. Differences in model physics, including the simulation distribution of precipitation, turbulent transport parameterizations, and parameterization of wet and dry removal, can also result in models  
5 simulating different concentrations, given the same emissions, so these results cannot necessarily be extended to other atmospheric models. In order to improve the original HS parameterization, an additional scaling factor of 6 has been added to the emission parameterization for our modelled concentrations to match the spore counts in Figure 2D. Although the correlation coefficient remains the same, this improves the MNMB and FGE metrics from 0.98 and 1.33 respectively to 0.18 and 1.00.

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Discrepancies between model and observations may be explained by an over-prediction of fungal spore sources via biases in the emission parameterization as formulated by (Hoose et al., 2010) or long-range transport, or an under-prediction of the rate of removal by dry and wet deposition. Additionally, as outlined by (Sesartic and Dallafior, 2011) and references therein, the observational data quality is limited and should be considered with caution. The methods used to measure actual spore  
15 concentrations may involve biases as well as problems related to the identification of fungal spores. As mentioned in section 2.3.1, (Sesartic and Dallafior, 2011) showed that many direct-counting spore techniques can significantly undercount spore number (i.e. by order of magnitude). Additionally, any culture-based methods have significant biases in that only a very small fraction of spore species can be culturable in a given medium.

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The total global emissions calculated here with HS ( $17 \text{ Tg yr}^{-1}$  corresponding to an average mass emission flux of  $2.5 \text{ ng m}^{-2} \text{ s}^{-1}$ ) are within the range of uncertainties reported by (Després et al., 2012) and (Fröhlich-Nowoisky et al., 2016). Using the same HS parameterization, (Heald and Spracklen, 2009) and (Hoose et al., 2010) calculated higher totals, respectively  $28 \text{ Tg yr}^{-1}$  and  $31 \text{ Tg yr}^{-1}$ . This demonstrates the model sensitivity to the leaf area index dataset used for that purpose and the specific humidity calculated by the model. The total global emissions calculated using SD and HU are estimated to  $86 \text{ Tg yr}^{-1}$  and  $349 \text{ Tg yr}^{-1}$ , respectively, which seem unrealistically high. Further, the comparison of fungal spore number fluxes  
25 calculated by (Sesartic and Dallafior, 2011) and by EMAC yields large discrepancies in magnitude and spatial distribution. This is most likely explained by large differences in the biome spatial distribution between MODIS and Olson data, leading to the higher emissions calculated by EMAC when using the SD parameterization. Since the HS simulation shows a better fit to observations, we will show results only from this simulation hereafter.

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Figure 3a shows that near-surface, annual mean number concentrations of fungal spores simulated by EMAC are typically  $2.6 \times 10^3 \text{ m}^{-3}$ , with a maximum over tropical forests of  $6 \times 10^3 \text{ m}^{-3}$ , matching the regions with largest emissions. These concentrations are one order of magnitude less than those calculated by (Heald and Spracklen, 2009) and (Spracklen and Heald, 2014) but within the range of concentrations reported by (Fröhlich-Nowoisky et al., 2009) and (Elbert et al., 2007).

The spatial distribution shows large similarities with other modeling studies mentioned above. Our simulated zonal annual mean number concentrations of fungal spores, presented in Figure 3b, decrease with altitude from a maximum at the Equator reaching up to  $4 \times 10^3$  to less than  $100 \text{ m}^{-3}$  at 250 hPa, contributing, in theory, very little to global CCN concentrations (Pringle et al., 2010b). This is beyond the scope of this study, and will be the focus of a follow-up paper.

5

### 3.2. Correlation between observations and meteorological parameters

Meteorological variables affect the initial release of fungal spores into the atmosphere and the dispersal once airborne. Temperature and humidity affect the size of the source and control the release of some actively released fungal spores (Jones and Harrison, 2004). Their frequency and concentrations are equally dependent on geographical characteristics. Since the publications collected for this study do not always provide information on the meteorological parameters of the observational site, we investigate the effects of physical parameters such as temperature, specific humidity and leaf area index as modeled by EMAC on the observed particle concentrations, taking into account the differentiation between ecosystems as defined by MODIS data. Interestingly, we found a strong difference between non-urban and urban observations through their correlations with the three parameters. Table 1 shows a correlation coefficient of 0.41 between the observations and specific humidity for the urban points and the same correlation between the non-urban observations and the leaf area index. This demonstrates the stronger effect of meteorological variables on urban sites, which could be taken into account in the formulation of a more advanced fungal spore emission parameterization. Additional observational evidence is needed to support this hypothesis.

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### 3.3. Bacteria concentrations

The total global emission source calculated by EMAC is  $0.76 \text{ Tg yr}^{-1}$ . This is within the range calculated by (Burrows et al., 2009a) ( $0.4 - 1.8 \text{ Tg yr}^{-1}$ ) with an older version of EMAC but using Olson vegetation types as ecosystem classification instead of the more recent, satellite-based MODIS vegetation distribution. Similar global emission estimates were reported by (Hoose et al., 2010) ( $0.7 \text{ Tg yr}^{-1}$ ). Although we calculate similar global annual mean number concentration ( $3.1 \times 10^4 \text{ m}^{-3}$ ) as in (Burrows et al., 2009a), we obtain differences in the geographical patterns and magnitudes of these concentrations in Figure 4a, most notably over tropical forest regions where the concentrations are much lower than over other continental regions. The differences can only be due to the replacement of Olson by MODIS data and the resulting spatial distribution of emissions. Our spatial distribution and magnitudes of number concentrations are very similar to those presented by (Spracklen and Heald, 2014), who also used MODIS land cover classifications.

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The zonal annual mean number concentrations of bacteria in Figure 4b are highest in the lower troposphere and decrease with altitude to reach about  $100 \text{ m}^{-3}$  at 100 hPa. This might have a slight impact on global CCN concentrations as

demonstrated by (Spracklen and Heald, 2014) as they found that bacteria contribute only 0.01% to global mean CCN concentrations near the surface, and that bacteria and fungal spores together contribute less than 1%.

A comparison of EMAC modeled PBAP concentrations (including bacteria, fungal spores and pollen) with FBAP observations from the IDEAS campaign in (Twohy et al., 2016) has shown that the model under-predicts observed concentrations in the free troposphere, while the most important PBAP category at high altitudes are bacteria, as indicated by Figure 4b.

### 3.4. Pollen concentrations

~~Figure 5a and 5b show the zonal, annual mean number concentration distributions of pollen near the surface.~~ The total global emission source calculated by EMAC is  $44 \text{ Tg y}^{-1}$ , and the average near-surface number concentration is  $24 \text{ m}^{-3}$ , being within the range of magnitudes reported by (Després et al., 2012) and references therein. ~~Surface annual mean number concentrations distribution and the zonal means are shown in the Supplementary Information.~~

### 3.5. Comparison of the EMAC simulated fungal spores and bacteria with FBAP observations

Figure 5 shows a comparison between the observed fluorescent particle number and the modeled concentrations of bacteria and fungal spores. ~~The comparison excludes data from Nanjing, China ( $\sim 2 \times 10^6 \text{ m}^{-3}$ ), because the observed particle counts were approximately two orders of magnitude higher than concentrations observed elsewhere, likely as a partial result of urban pollution.~~ For the locations Saclay and Killaney, both UV-APS and WIBS observations are available, though, for the sake of clarity, only WIBS data are shown in Figure 5.

In general, the model results compare relatively well with the observations of total fluorescent particles in almost all locations. The model concentrations are lower than FBAP concentrations in ca. 40% of the observations and are within a factor of two (high or low) in all cases. This model under-prediction is best observed in the summer season. The model predicts higher bacteria loadings than fungal spores (including in Nanjing) in almost all cases, with the only exceptions of Ucluelet (rural coastal) and the Amazon (near-pristine forest). Additionally, the simulated bacteria number concentrations in Figure 5 dominate the total PBAP concentrations for the locations where observations are available in winter, when very low concentrations of fungal spores are expected. A possible explanation for this discrepancy is that bacteria emissions are assumed constant in time, representing inferred “background” emissions, while they may exhibit seasonal variations not taken into account in our model. Consequently, the differences in the seasonal bacteria concentrations for each location are related to transport and deposition patterns reproduced in the model. Fungal spore emissions, in contrast, are assumed to include seasonal variability due to the seasonality of the leaf area index and the specific humidity, which are used as model inputs. Many reports support the observed seasonal cycle of fungal spore concentrations, which are typically highest in

summer and early fall, but depends on latitude and ecosystem (Lacey, 1996; Lang-Yona et al., 2012; Manninen et al., 2014; Schumacher et al., 2013). Laboratory-based observations have shown that the WIBS FL3 channel utilized here is less efficient at detecting bacteria (Hernandez et al., 2016; Savage et al., 2017) and thus is likely to detect fungal spores or pollen with relatively higher efficiency. The large size of pollen grains limits their ability to be detected by the WIBS, however, and so fungal spores are assumed to represent the largest fraction of biological particles detected by the FL3 WIBS channel used here. Moreover, evidence in certain campaigns suggests that these fluorescence channels correlate well with fungal spores (e.g., (Fernandez-Rodriguez et al., 2018; Healy et al., 2014; Huffman et al., 2012)). Other important technical considerations were discussed in Section 2.3.2.

10 The simulated fungal spores show the lowest concentrations during the winter season, and can be significant in the summer or dry seasons. This is especially the case for the Amazon and Ucluelet stations and in Borneo, where fungal spores were shown by the model to dominate the total PBAP concentrations (Fig. 5). If we consider that the FBAPs observed in these locations mainly represent fungal spores, the model underestimates the fungal spores concentrations. This contradicts results presented in Section 3.1, which shows a modeled overestimation with respect to spore counts measured by optical  
15 microscopy (Fig. 2). As mentioned, the comparison of modeled spore results with direct counts of spores may also be biased due to model inputs and because of the frequently observed undercounting of some collection and detection methods used for spore counting. FBAP and spore counts measured via optical microscopy were compared for two sets of new measurements shown here. The observed values from each method are reported here with the relative factor that the FBAP concentration overcounts the spore counts from the optical technique shown in parentheses: Saclay -  $0.048 \times 10^6 \text{ m}^{-3}$  (spore count),  $0.088 \times 10^6 \text{ m}^{-3}$  (WIBS-4A; x1.8) and  $0.027 \times 10^6 \text{ m}^{-3}$  (UV-APS; x0.6); Cyprus -  $0.0015 \times 10^6 \text{ m}^{-3}$  (spore count),  $0.0433 \times 10^6 \text{ m}^{-3}$  (WIBS-4A; x29). In several other cases, collocated measurements of FBAP and spore counts also show spore count to be lower than FBAP by a factor of  $\sim 2$  (Fernandez-Rodriguez et al., 2018) to as  $\sim 10$  (Healy et al., 2014; Huffman et al., 2012), again depending partially on instrumental parameters and differences in aerosols observed. Additional direct comparison of FBAP concentrations with spore count concentrations summarized in this study (Table S11 and Fig. 2)  
20 is not possible because of differences in locations and seasons. Therefore, the qualitative relationship between UV-LIF and other spore counting techniques has been demonstrated, but quantitative comparisons often show significant differences.

Significant uncertainties also still remain in the interpretation of the UV-LIF measurements. For example, UV-LIF measurements do not detect all spore types equally. (Hernandez et al., 2016; Savage et al., 2017) show differences in  
30 fluorescence profiles of a number of spore types and discuss that different instrument units detect particles with different detection efficiencies. This implies that both biological and instrumental factors can lead to differences in observed FBAP concentrations. (Healy et al., 2014; Huffman et al., 2012) both discuss how certain types of spores may escape detection. In particular, (Fernandez-Rodriguez et al., 2018; Healy et al., 2014) discuss how the genus *Cladosporium* (among the most commonly observed spore type in many environments) correlated very poorly with fluorescent measurements, suggesting

that dark-walled cell walls present in this type of spores may inhibit some types of real-time fluorescence detection. This spore type is a dominant spore type during dry weather, therefore it might be undercounted during the day and in certain locations where such spore types are a high fraction of the spore number. In some cases the FL3 signal will also be influenced by non-biological particles (Savage et al., 2017), and so FBAP number concentrations from WIBS and UV-APS presented here should be used only for rough comparison.

### 3.6. Chemical aerosol mass composition simulated by EMAC

To evaluate the presence of PBAP particles (including bacteria, fungal spores and pollen) relative to other particle types, we perform a comparison to the dry aerosol composition simulated by EMAC, including dust, organic carbon, black carbon, sea salt and inorganic anthropogenic aerosols. The PBAP mass concentration **at the surface** is estimated from the number concentration assuming monodisperse and spherical particles (see Section 2) with a mass per particle of 0.52 pg (Burrows et al., 2009a), 33 pg (Fröhlich-Nowoisky et al., 2016) and 250 ng (Miller, 1982) for bacteria, fungal spores and pollen, respectively. The model calculated mean total global aerosol mass concentration is approximately  $65 \mu\text{g m}^{-3}$ . Global mean PBAP mass concentrations represent a total mass of approximately  $5 \mu\text{g m}^{-3}$ , being dominated by pollen, which is less than 7% of the total simulated mass concentration. Desert dust is the main contributor to the total mass, as shown in figure 6A. When dust is excluded (Figure 6B) PBAPs account for 19% of the total mass concentration, while their contribution is 52% when both dust and sea salt are excluded (Figure 6C) Under the first assumption and considering that dust is mostly concentrated over deserts, fungal spores and pollen mass concentrations contribute significantly to the total mass concentrations in central Africa (6% and 14%, respectively) and South America (5% and 11%, respectively). ~~These values are much lower than the number and mass fractions of 80% reported by (Pöschl et al., 2010).~~

## 4. Discussion and Conclusions

We used the chemistry-climate model EMAC to simulate the total PBAPs present in the atmosphere including bacteria, fungal spores, and pollen. We compared the simulated fungal spore number concentrations, using three emission parameterizations, to an updated data set of spore counts synthesized from the literature. Additional evaluation of the model simulated PBAP concentrations was performed using a comparison with a global data set of FBAP concentrations.

Bacteria were assumed to have an emission diameter of  $4 \mu\text{m}$ , based on observations of bacteria-containing particles and clusters of bacteria. This allowed the inclusion of bacteria loadings in our model comparison with FBAP observations. Sensitivity runs with lower emission diameters ( $1 \mu\text{m}$  and  $2 \mu\text{m}$ ) showed similar near-surface global mean concentrations. The global distribution of bacteria concentrations is therefore not expected to change by varying the diameter.



From three fungal spore emission schemes, the HS parameterization showed the best fit to observations, although the model over-predicts the concentrations by up to a factor of 6 in some locations. Spore count observations were limited in time and space and are subject to several methodological issues. Therefore, these direct-count measurements cannot provide a rigorous evaluation of the model results. Bacteria and fungal spore concentrations predicted by the model compare well with FBAP observations from real-time measurement techniques. The overprediction/underprediction is estimated to be of a factor of 2 or less for all measurement locations. Modeled bacteria concentrations outnumber fungal spores in most locations, particularly in winter, while simulated fungal spores might be underestimated in near-pristine environments. The FBAP concentrations used here (from UV-APS data and only FL3 for WIBS) are likely to underestimate fungal spores somewhat and dramatically underrate the abundance of bacteria in most locations. Most bacteria are not strongly fluorescent in the applied wavelength channels and are therefore underestimated. This might explain the differences when we expect high bacterial counts (e.g. (Savage et al., 2017)).

We conclude that fungal spore concentrations are underestimated by the model in most locations with respect to the FBAP data used, but overestimated with respect to direct count observations of spores. Furthermore, the bacteria emission scheme we applied does not represent seasonal nor diurnal variability, a weakness that needs to be addressed in future work. Further comparisons of our model results with long-term FBAP measurements, taking into account daily and seasonal variability, might offer new opportunities to better constraint our model emission parameterizations. Improvements in the way FBAP data are analyzed will also allow for better separation of bacteria and fungal spores and will thus allow for better comparison with improved models in the future (Ruske et al., 2017).

We find that PBAPs contribute up to 19% to the total aerosol mass concentration globally after excluding dust aerosols. This percentage is dominated primarily by pollen. Regionally, fungal spores may contribute significantly to the total aerosol mass. PBAP, fungal spores and pollen especially, account for a major part of the aerosol loading and are likely underestimated by our model. In the future we anticipate further adding an evaluation of future updates to the model, **using the new pollen emission model for climate models of (Wozniak and Steiner, 2017)**, with respect to modeled pollen concentrations in order to better characterize total PBAP mass.

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	Specific humidity	Temperature	Leaf Area Index
Urban Observations	<b>0.41</b>	0.26	-0.07
Non-Urban Observations	0.02	0.15	<b>0.41</b>

Table 1: Correlation table between observations and the modeled specific humidity, temperature and leaf area index. The values in bold are statistically significant ( $p < 0.05$ ).

Location	LON	LAT	ALT	Instrument	Start	End	Season	Site category	Publication	Size Range ( $\mu\text{m}$ )	Mean ( $10^6\text{m}^{-3}$ )
Amazon, Brazil	-60,2	-2,58	110	UV-APS	03.02.2008	15.03.2008	Winter	rainforest	(Huffman et al., 2012)	>1	0,093
Karlsruhe, Germany	8,42	49,09	117	WIBS-4A	01.04.2010	01.04.2011	Yearly	semi-rural	(Toprak and Schnaiter, 2013)	0.8 -16	0,031
Karlsruhe, Germany	8,42	49,09	117	WIBS-4A	01.04.2010	01.07.2010	Spring	semi-rural	(Toprak and Schnaiter, 2013)	0.8 -16	0,029
Karlsruhe, Germany	8,42	49,09	117	WIBS-4A	01.07.2010	01.10.2010	Summer	semi-rural	(Toprak and Schnaiter, 2013)	0.8 -16	0,046
Karlsruhe, Germany	8,42	49,09	117	WIBS-4A	01.10.2010	01.01.2011	Autumn	semi-rural	(Toprak and Schnaiter, 2013)	0.8 -16	0,029
Karlsruhe, Germany	8,42	49,09	117	WIBS-4A	01.01.2011	01.04.2011	Winter	semi-rural	(Toprak and Schnaiter, 2013)	0.8 -16	0,019
Colorado, USA	-105,15	39,16	2290	UV-APS	20.07.2011	22.08.2012	Yearly	rural	(Schumacher et al., 2013)	>1	
Colorado, USA	-105,15	39,16	2290	UV-APS	01.03	31.05	Spring	rural	(Schumacher et al., 2013)	>1	0,015
Colorado, USA	-105,15	39,16	2290	UV-APS	01.06	31.08	Summer	rural	(Schumacher et al., 2013)	>1	0,03
Colorado, USA	-105,15	39,16	2290	UV-APS	01.09	30.11	Autumn	rural	(Schumacher et al., 2013)	>1	0,017
Colorado, USA	-105,15	39,16	2290	UV-APS	01.12	29.02	Winter	rural	(Schumacher et al., 2013)	>1	0,0053
Hyytiälä, Finland	24,17	61,85	181	UV-APS	27.08.2009	17.04.2011	Yearly	boreal forest	(Schumacher et al., 2013))	>1	
Hyytiälä, Finland	24,17	61,85	181	UV-APS	01.03	31.05	Spring	boreal forest	(Schumacher et al., 2013)	>1	0,015
Hyytiälä, Finland	24,17	61,85	181	UV-APS	01.06	31.08	Summer	boreal forest	(Schumacher et al., 2013)	>1	0,046
Hyytiälä, Finland	24,17	61,85	181	UV-APS	01.09	30.11	Autumn	boreal forest	(Schumacher et al., 2013)	>1	0,027
Hyytiälä, Finland	24,17	61,85	181	UV-APS	01.12	29.02	Winter	boreal forest	(Schumacher et al., 2013)	>1	0,004
Killaney, Ireland	-9,5	52,05	34	WIBS-4A	02.08.2010	02.09.2010	Summer	rural	(Healy et al., 2014)	>1	0,035
Killaney, Ireland	-9,5	52,05	34	UV-APS	02.08.2010	02.09.2010	Summer	rural	(Healy et al., 2014)	>1	0,015
Mainz, Germany	8,23	49,98	100	UV-APS	03.08.2006	04.12.2006	Autumn	semi-urban	(Huffman et al., 2010)	>1	0,027
Borneo, Indonesia	117,84	4,98		WIBS-3	01.06.2008	31.07.2008	Summer	forest	(Gabey et al., 2010)	0.8-20	0,2
Nanjing, China	118,95	32,12		WIBS-4A	29.10.2013	15.11.2013	Autumn	urban	(Yu et al., 2013)	>1	2,09
Puy de dôme, France	2,96	45,43	1465	WIBS-3	22.06.2010	03.07.2010	Summer	mountain	(Gabey et al., 2013)	>1	0,095
Manchester, UK	-2,25	53,48		WIBS-3	04.12.2009	21.12.2009	Winter	urban	(Gabey et al., 2011)	0.8-20	0,11



Helsinki, Finland	24,65	60,2		UV-APS	02.02.2012	25.02.2012	Winter	suburban	(Saari et al., 2015)	<1	NA
Helsinki, Finland	24,65	60,2		UV-APS	16.06.2012	22.08.2012	Summer	urban	(Saari et al., 2015)	>1	0,013
Helsinki, Finland	24,65	60,2		BIO-SCOUT	02.02.2012	25.02.2012	Winter	suburban	(Saari et al., 2015)	>1	0,01
Helsinki, Finland	24,65	60,2		BIO-SCOUT	16.06.2012	22.08.2012	Summer	urban	(Saari et al., 2015)	>1	0,028
Munnar, India	77,06	10,09	1605	UV-APS	01.06.2014	20.08.2014	Summer	rural	(Valsan et al., 2016)	>1	0,017
Ucluelet, Canada	-125,54	48,92	5	WIBS-4A	06.08.2013	28.08.2013	Summer	rural/marine	unpublished	>1	0,059
Paris, France	2,35	48,85		UV-APS	14.01.2010	15.02.2010	Winter	urban	unpublished	>0.8	0,0276
Saclay, France	2,17	48,71		UV-APS	16.06.2014	05.08.2014	Summer	semi-urban	unpublished	>0.8	0,027
Saclay, France	2,17	48,71		WIBS-4A	16.06.2014	05.08.2014	Summer	semi-urban	unpublished	>0.8	0,088
Cyprus	33,06	35,03	550	WIBS-4A	01.04.2016	26.04.2016	Spring	rural/mountain	unpublished	>0.8	0,0433
Barbados	-59,43	13,16	5	WIBS-4A	16.07.2016	04.09.2016	Summer	rural/marine	unpublished	>0.8	0,0951

Table 2: List of the FBAP observations.

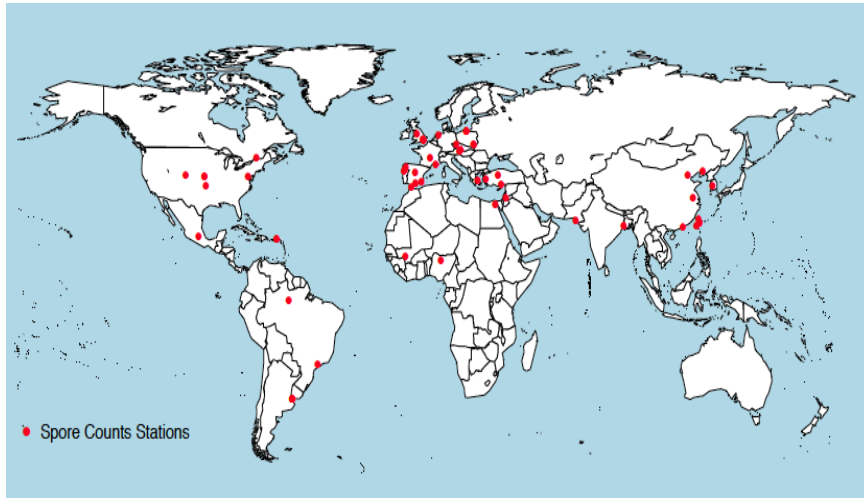


Fig. 1: Geographical locations of the fungal spore counts (List of locations is provided in Table S11).

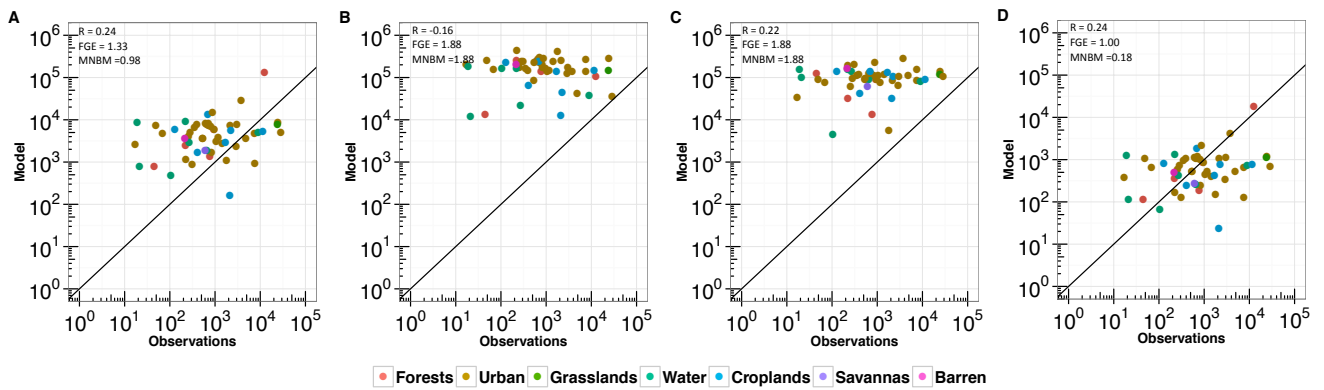


Fig. 2: Comparison between fungal spores number concentrations observed and simulated by EMAC using three emission parameterizations: A. (Heald and Spracklen, 2009), B. (Sesartic and Dallafior, 2011), C. (Hummel et al., 2015) and D. (Heald and Spracklen, 2009), scaled by a factor of 6. Point Colors depict the ecosystems of the observational stations as defined by MODIS. Units:  $m^{-3}$ .

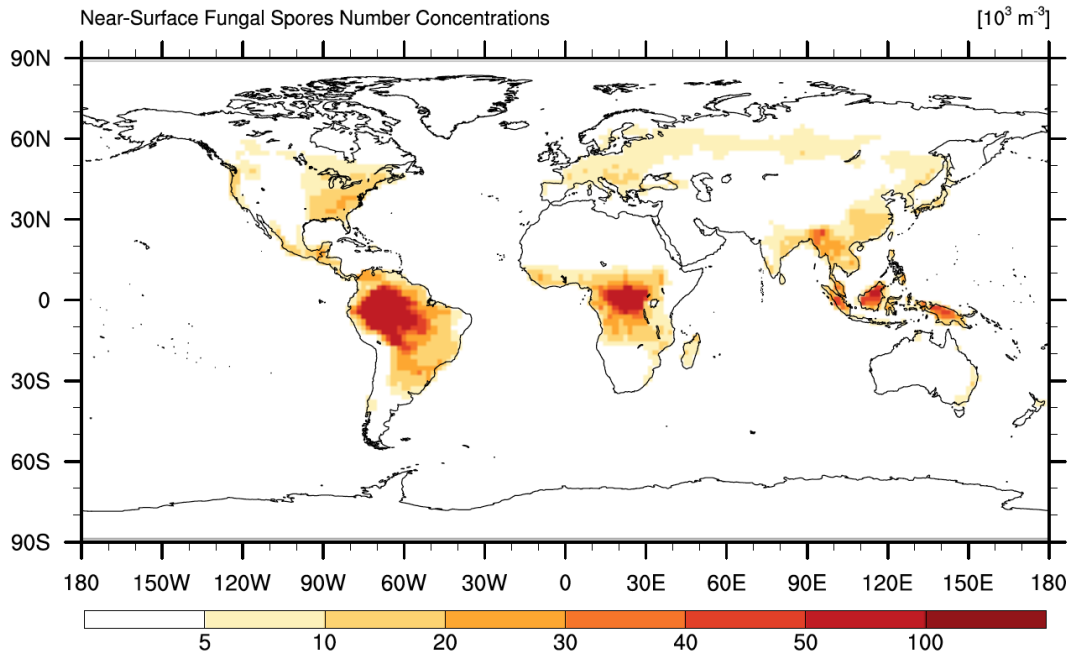


Fig. 3a: Modeled near-surface annual mean number concentration of fungal spores (in 10<sup>3</sup> m<sup>-3</sup>).

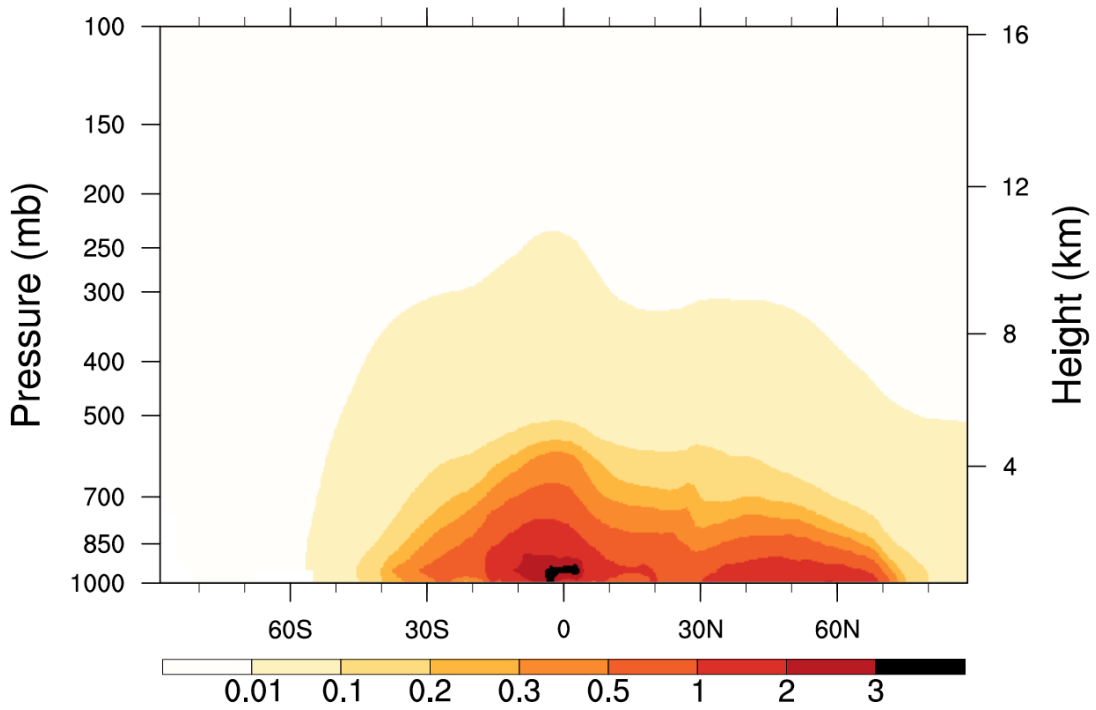


Fig. 3b: Zonal annual mean number concentrations of fungal spores (in 10<sup>3</sup> m<sup>-3</sup>).

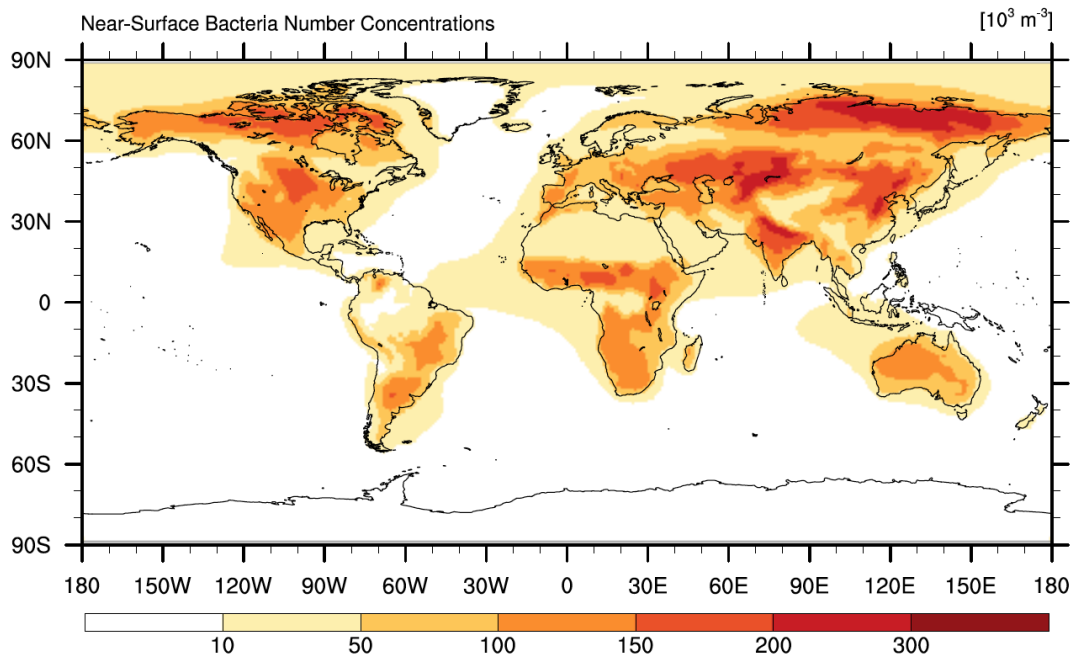


Fig. 4a: Modeled near-surface annual mean number concentration of bacteria (in  $10^3 \text{ m}^{-3}$ ).

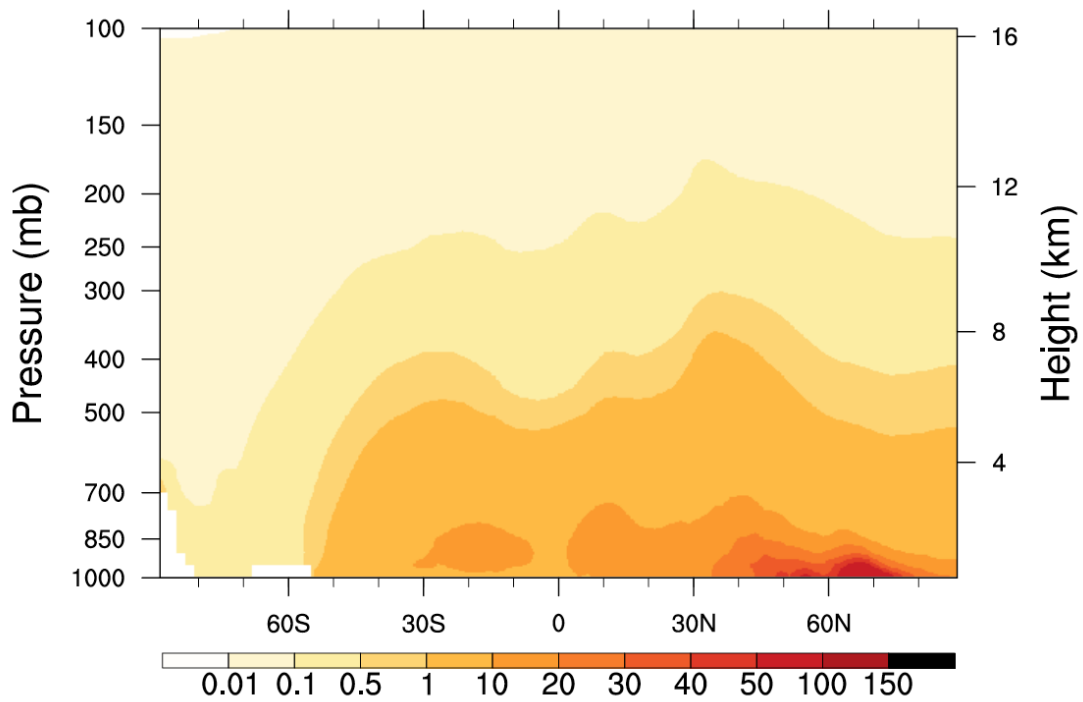


Fig. 4b: Zonal annual mean number concentrations of bacteria (in  $10^3 \text{ m}^{-3}$ ).

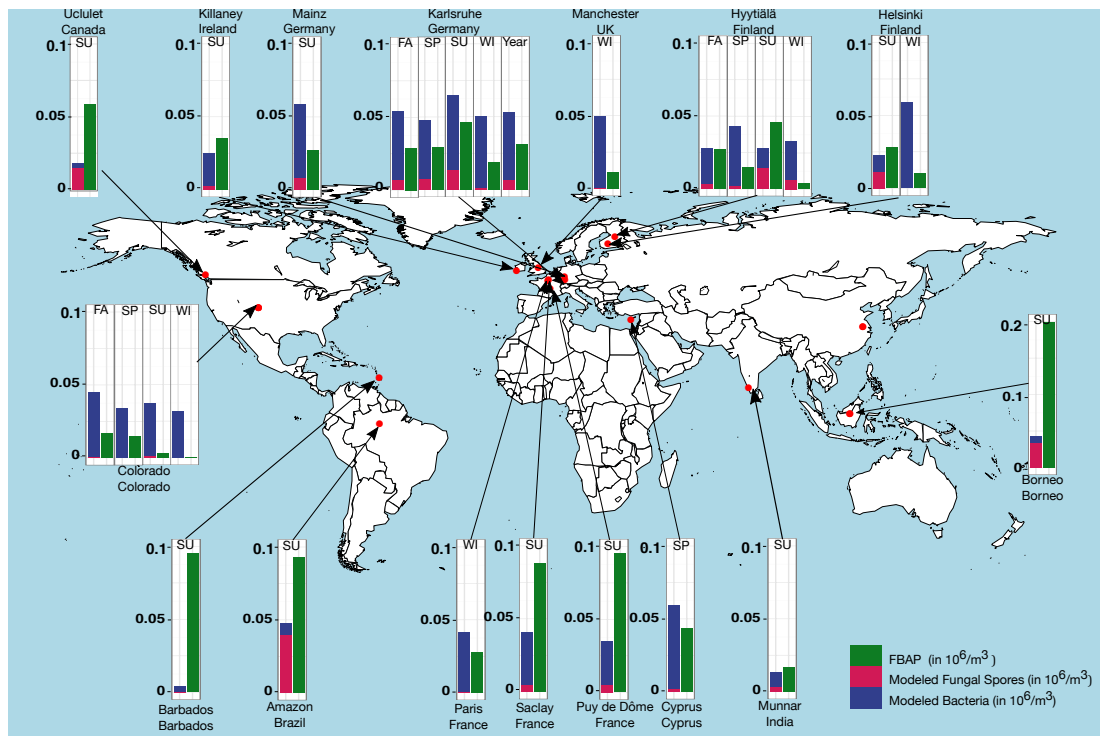
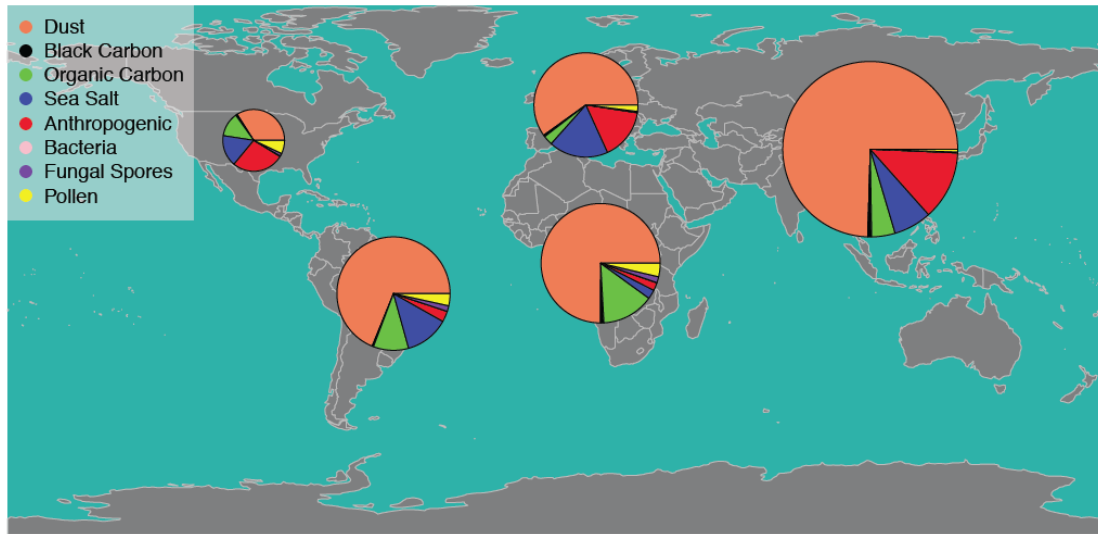
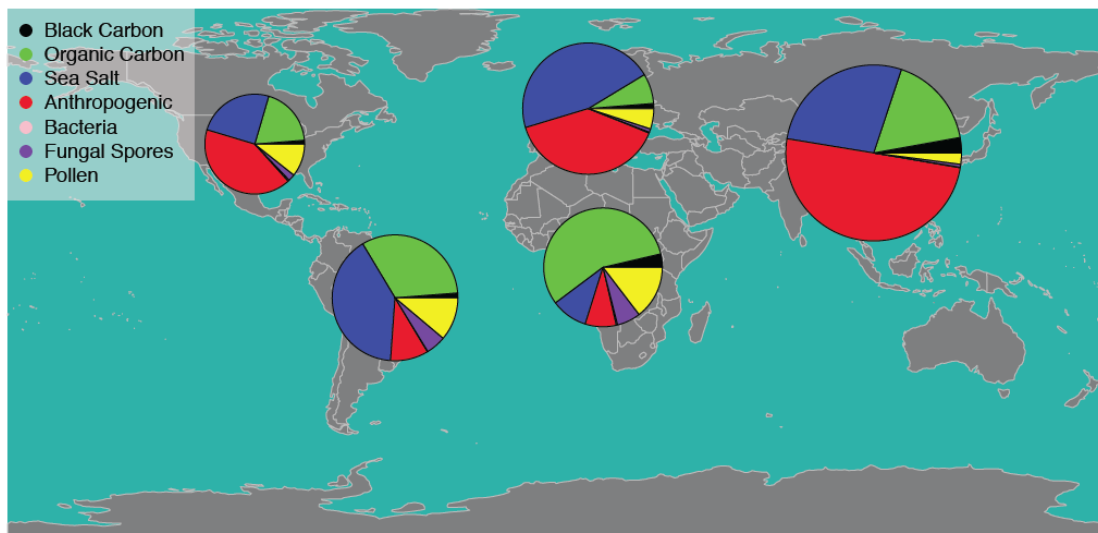


Fig. 5: Comparison between observed FBAP and modeled bacteria and fungal spores sampled for the campaigns described in Table SI 2. Units:  $10^6 m^{-3}$ . Seasons of observations are: FA (Fall), SP (Spring), WI (Winter) and Year for yearly observations. Data from Borneo are plotted on a different scale because of the high particle count observations and data from Nanjing, China were removed here due to heavy urban influence.

A



B



C

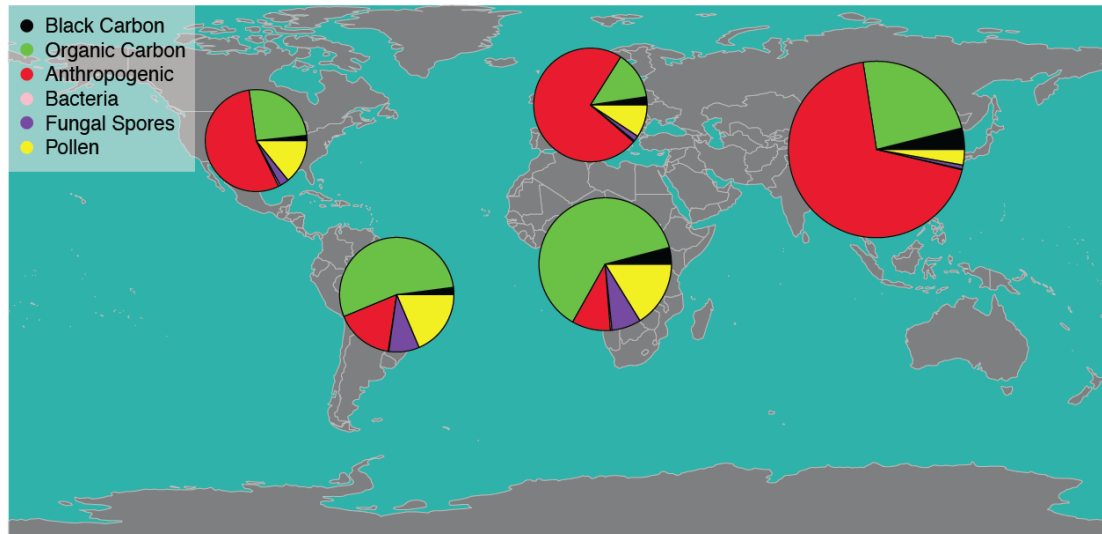


Figure 6: Near-surface annual mean chemical aerosol mass composition simulated by EMAC for five regions: North America ( $126^{\circ}\text{W} - 72^{\circ}\text{W}/30^{\circ}\text{N} - 52^{\circ}\text{N}$ ), Europe ( $12^{\circ}\text{W} - 36^{\circ}\text{E}/34^{\circ}\text{N} - 62^{\circ}\text{N}$ ), East Asia ( $100^{\circ} - 144^{\circ}\text{E}/20^{\circ}\text{N} - 44^{\circ}\text{N}$ ), Central Africa ( $10^{\circ}\text{E} - 40^{\circ}\text{E}/10^{\circ}\text{S} - 10^{\circ}\text{N}$ ) and South America ( $75^{\circ}\text{W} - 35^{\circ}\text{W}/30^{\circ}\text{S} - 0^{\circ}\text{N}$ ). A. with dust, B. without dust, C. without dust and sea salt. The size of the pie chart is proportional to the total aerosol mass concentration.

## Supplementary Information

Table SI1. Bacterial emission fluxes (best-fit) estimated by Burrows et al. (2009a).

<b>Ecosystem</b>	<b>Best-fit <math>F_i</math> (<math>m^{-2}s^{-1}</math>)</b>
Coastal	900
Crops	700
Deserts	0
Forests	0
Grasslands	648
Land-Ice	0
Sea	0
Shrubs	502
Tundra	0
Wetlands	196



**Table SI2: List of the fungal spores counts (in m<sup>-3</sup>)**

Reference	LON	LAT	ALT	startdate	enddate	Mean	Min	Max	Reference
AM05	2,95	45,76	1465	04.03.2003	06.03.2003	222	53	390	(Amato et al., 2005)
BA02	15,76	47,71	1644	01.03.2000	30.03.2000	773	104	2031	(Bauer et al., 2002)
BE85	6,6	53,16	5	01.01.1981	31.12.1983	258	42	1225	(Beaumont et al., 1985)
CO08	-73,75	45,46	0	16.10.2002	16.10.2002	615	492	738	(Côté et al., 2008)
DA63_1	-0,11	51,5	64	01.06.1961	31.10.1961	7500	2000	13000	(Davies et al., 1963)
DA63_2	-2,98	53,4	92	01.06.1961	31.10.1961	9000	3000	15000	(Davies et al., 1963)
DIG96	5,36	43,28	0	01.01.1962	31.12.1962	69	3	411	(Di Giorgio et al., 1996)
EL07	-59,4	-1,91	174	21.07.2001	21.07.2001	12476	4764	20188	(Elbert et al., 2007)
FI90	14,41	50,08	0	01.10.1988	15.12.1988	17	NA	NA	(Fišar et al., 1990)
DR66	7,63	11,18	690	01.07.1959	30.07.1959	128	20	237	(Dransfield, 1966)
GON10	-46,63	-23,55	0	01.03.2007	31.03.2007	3768	NA	NA	(Goncalves et al., 2010)
BARD13	-5,36	35,56	250	01.01.2009	31.12.2012	1434	177	12082	(Bardei et al., 2013)
KAS_1	22	50,03	0	01.01.2001	31.12.2002	2163	NA	NA	(Kasprzyk and Worek, 2006)
KAS_2	22,08	50,03	0	01.04.2001	31.12.2002	2119	NA	NA	(Kasprzyk and Worek, 2006)
MA01_1	18,8	54,6	0	01.01.1998	31.03.1998	105	0	1000	(Marks et al., 2001)
MA01_2	18,8	54,6	0	01.07.1998	30.09.1998	223	0	600	(Marks et al., 2001)
MA01_3	18,85	54,52	0	02.08.1995	17.09.1995	19	0	122	(Marks et al., 2001)
OLI09_1	-8,38	41,51	5	01.01.2005	31.12.2007	934	NA	NA	(Oliveira et al., 2009)
OLI09_2	-8,65	41,18	20	01.01.2005	31.12.2007	531	NA	NA	(Oliveira et al., 2009)
RO05_2	-7,85	42,33	0	01.01.2002	31.12.2002	964	NA	NA	(Rodríguez-Rajo et al., 2005)
GRIF_01	-64,73	18,33	0	01.07.2000	30.07.2000	45	30	60	(Griffin et al., 2001)
GRIF_03	-64,79	18,33	0	18.07.2000	08.08.2001	21,24	18,25	61	(Griffin et al., 2003)
GRIF_07	34,25	36,6	0	18.03.2002	22.10.2002	1702	100	8510	(Griffin et al., 2007)
LAU06	114,26	22,33	0	01.09.2002	30.04.2003	269,5	28,5	1963	(Lau et al., 2006)
KAT97	-104,86	39,75	0	01.01.1987	31.12.1995	409	NA	NA	(Katial et al., 1997)
PAD57_1	-96,56	39,18	0	01.04.1953	30.10.1953	24499	837	48162	(Pady, 1957)
PAD57_2	-96,56	39,18	0	01.12.1953	30.03.1954	715	170	1261	(Pady, 1957)
LEV06	-95,93	36,11	0	01.01.2002	31.12.2002	24121	53	48188	(Levetin and Dorsey, 2006)
MAL10_1	-57,95	-34,91	15	01.07.2000	15.09.2000	284	NA	2000	(Mallo et al., 2010)
MAL10_2	-57,95	-34,91	15	15.09.2000	15.12.2000	395	NA	2000	(Mallo et al., 2010)
MAL10_3	-57,95	-34,91	15	15.12.2000	15.03.2001	814	NA	3478	(Mallo et al., 2010)
MAL10_4	-57,95	-34,91	15	15.03.2001	30.06.2001	715	NA	4763	(Mallo et al., 2010)
SAB00	-3,58	37,18	0	01.01.1994	31.12.1994	832	NA	NA	(Sabariego et al., 2000)
SAK03	32,86	39,93	15	01.01.1990	31.12.1990	2917	17	5817	(Şakiyan and Inceoğlu, 2003)
HAM59	-0,35	51,81	0	04.05.1954	30.09.1954	11500	6400	10000	(Hamilton, 1959)

WU07	121,1	25,05	0	01.03.2003	31.12.2004	2255	NA	NA	(Wu et al., 2007)
WU04	120,2	23	0	01.12.2000	30.04.2001	28683	NA	NA	(Wu et al., 2004)
KE04	-8	12,65	0	01.02.2001	31.03.2001	225	80	370	(Kellogg et al., 2004)
CHO97	127,38	36,35	0	01.01.1995	31.12.1995	3014	100	5929	(Choi et al., 1999)
HO15	121,6	23,96	0	01.04.1993	31.03.1996	4839	NA	NA	(Ho et al., 2005)
AB14	36,1	32,01	0	01.12.2008	30.09.2009	7541	3066	12017	(Abu-Dieyeh et al., 2010)
PY14	23,71	37,98	30	01.01.1998	31.12.2001	1055	60	6328	(Pyrri and Kapsanaki-Gotsi, 2015)
RO90	-99,18	19,31	0	01.08.1988	28.02.1989	351	45	3195	(Rosas et al., 1990)
EL13	-0,98	37,6	0	01.01.1994	31.12.1999	655	10	1301	(Elvira-Rendueles et al., 2013)
HAS12	67	24,85	0	01.01.2010	31.12.2010	310	157	469	(Hasnain et al., 2012)
HE06	-3,75	40,45	0	01.01.2003	31.12.2003	609	166	1614	(Herrero et al., 2006)
WI09	16,36	48,2	0	01.01.2002	31.12.2002	49	NA	NA	(Winiwarter et al., 2009)
ABD12	31,55	29,86	0	01.01.2006	29.02.2007	216	101	331	(Abdel Hameed et al., 2012)
HU05	116,38	39,91	0	01.06.2003	30.05.2004	1164	23	13959	(Fang et al., 2005)
ZHA00	118,76	32,05	0	21.03.1998	14.07.1998	655	NA	NA	(Zhai et al., 2000)
HU94	123,06	41,13	0	01.12.1990	30.11.1991	1797	NA	NA	(Hu et al., 1994)
JO83	-77,33	39,23	0	01.09.1978	01.12.1980	695	4	6885	(Jones and Harrison, 2004)
KA10	27,41	38,61	74	01.01.2004	31.12.2005	541	415	780	(Kalyoncu, 2010)
AD03	88,16	22,66	10	01.10.1996	30.09.1998	879	119	1639	(Adhikari et al., 2004)

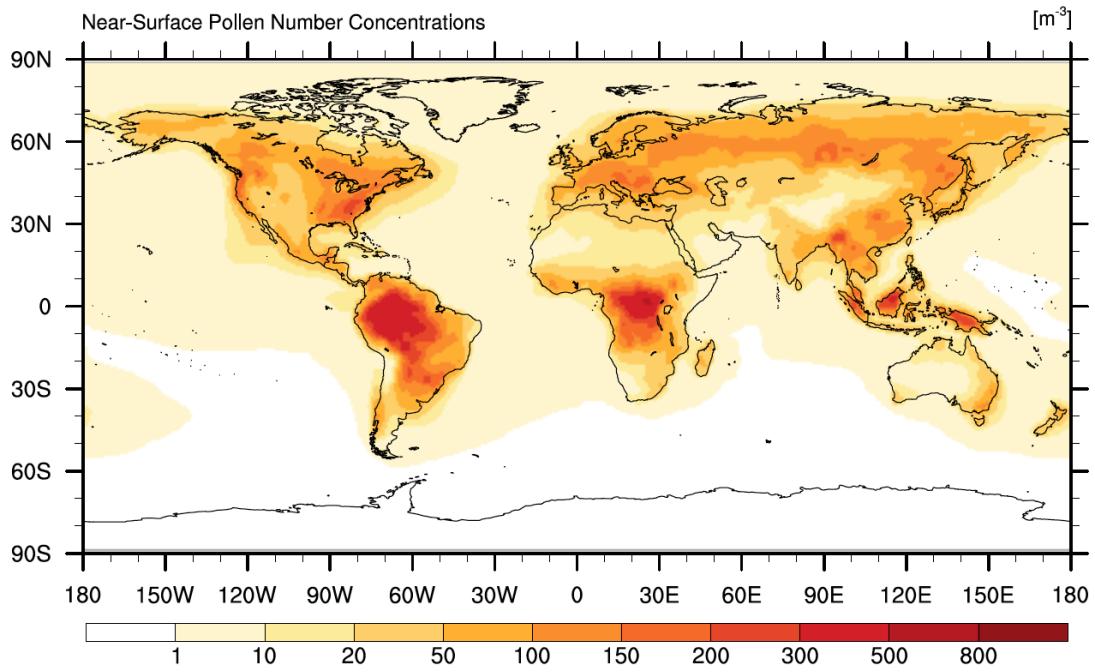


Fig. SIa: Modeled near-surface annual mean number concentration of pollen (in m<sup>-3</sup>).

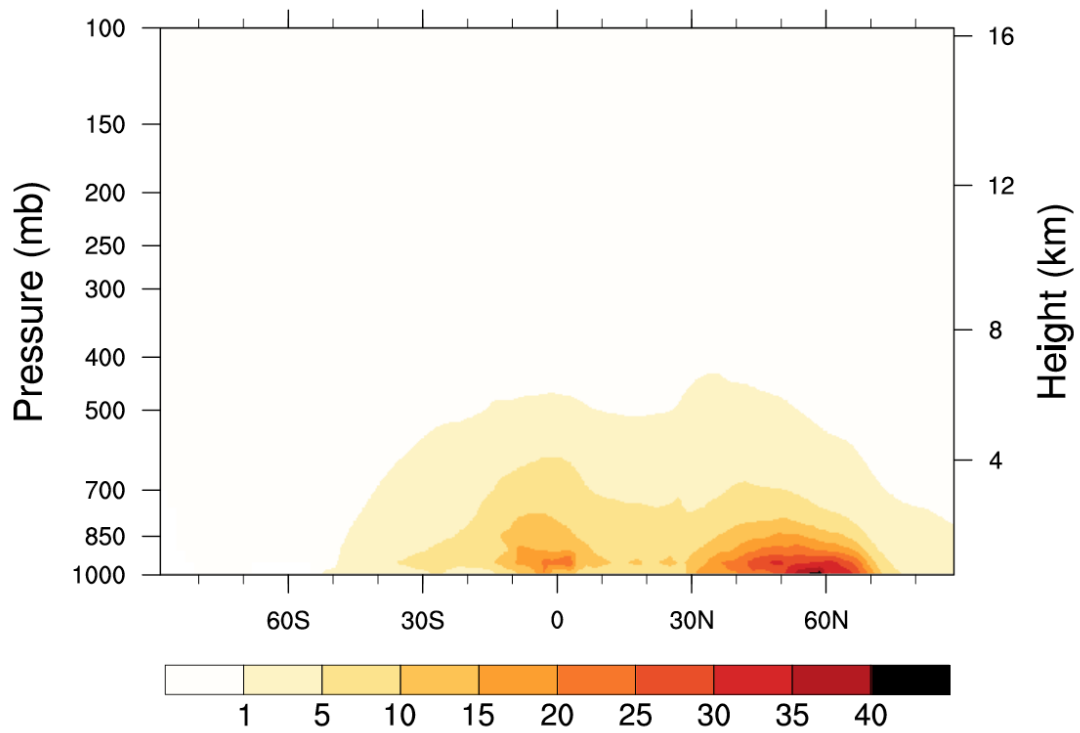


Fig. SIb: Zonal annual mean number concentrations of pollen (in m<sup>-3</sup>)

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