Interactive comment on "Regional-scale Simulations of Fungal Spore Aerosols Using an Emission Parameterization Adapted to Local Measurements of Fluorescent Biological Aerosol Particles" by M. Hummel et al.

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We thank the reviewer for his/her comments which have improved our manuscript. The comments are listed below in italics.

There is insufficient statistical support for the arguments put forth in this study. The authors need to fully quantify model performance for each scheme (R=correlation coefficient and normalized mean bias statistics for Figure 3-7). In doing so, it is critical that the authors distinguish between bias and skill. Does the new scheme actually add to the model skill (i.e. improve R2) or simply eliminate bias? Are there other potential causes for the bias (i.e. LAI, qv, constants used in the model)?

In order to improve the statistical support for this study's arguments further information on model performance is included in this reply and will be added to the manuscript. The following content to this comment is also added to the manuscript. For each time series, a correlation coefficient (R^2) and a normalized mean bias (NMB; Im et al., 2013) have been calculated by

$$R^2 = 1 - \frac{\sum_{i=1}^{n}\left(M_i - \widehat{O_i}\right)^2}{\sum_{i=1}^{n}(M_i - \bar{M})^2}$$

$$NMB = \frac{\sum_{i=1}^{n}(M_i - O_i)}{\sum_{i=1}^{n} O_i}\ 100$$

which gives an indication for a correlation between simulated fungal spore concentrations (M: $N_{H\&S}$, $N_{S\&D}$, and N_{FBAP}) and measured FBAP concentrations (O: $N_{F,C}$). Index i represents single elements of each time series. Elements calculated by a linear regression between simulated and measured concentrations are labeled with a hat, the mean value of a time series is labeled with an overbar. Results are listed in Table 1. Please note that the bottom line in Table 1 is not representing a mean value of the numbers above, but represents the result for a combination of all the time series. As noted by referee, different behaviors are found for R^2 and NMB and therefore it is important to distinguish between skill and bias in further discussion.

Table 1. Correlation coefficient (R^2) and normalized mean bias (NMB) for correlations between fungal spore and FBAP concentrations at different locations and three different time periods

	$N_{H\&S}$		$N_{S\&D}$		N_{FBAP}	
	R^2	*NMB*	*R^2*	*NMB*	*R^2*	*NMB*
Karlsruhe, Jul10	0.0047	-57.54	0.0127	-66.30	0.0048	-31.86
Karlsruhe, Aug10	0.0125	-63.82	0.0002	20.14	0.0274	-34.68
Karlsruhe, Oct10	0.0125	-35.71	0.0052	-40.08	0.0124	1.36
Hyytiälä, Jul10	0.3268	3.18	0.3358	-67.49	0.3255	50.50
Hyytiälä, Aug10	0.0002	-57.54	0.0099	35.43	0.0511	-59.04
Hyytiälä, Oct10	0.0016	-61.74	0.0350	-75.56	0.0000	-45.97
Manchester, Aug10	0.4558	2.74	0.4274	37.72	0.4409	63.26
Killarney, Aug10	0.2408	84.42	0.0998	200.51	0.1982	213.81
all	**0.1834**	**-44.04**	**0.0551**	**-28.74**	**0.1877**	**-0.43**

The statistical analysis of the results indicates that NMB improves more than R^2. Differences in R^2 are especially small between $N_{H\&S}$ and N_{FBAP}, because both make use of emission rates as a function of almost the same parameters (N_{FBAP} includes an additional T-dependence). Parameters b_1 and b_2 (in eq. 8 of the manuscript) are estimated to give fungal spore concentrations matching best with measured FBAP concentrations. At T = 275.82 K, $F_{H\&S}$ is equal to F_{FBAP}, and temperatures above this threshold (as it is the case for almost all locations) shift F_{FBAP} to give a larger emission flux. At meteorological conditions present for the selected cases, the second part of eq. 8 dominates over the first part by a factor of ~4 and therefore temperature changes have only a secondary influence on the emission flux. Hence, R^2 is similar for both emission parameterizations.

Possible causes for the bias of $F_{H\&S}$ and $F_{S\&D}$ may come from different assumptions made to determine the fungal spore concentrations in ambient air. The mass size distribution of mannitol, which is used as a chemical tracer for fungal spores by Heald and Spracklen (2009), peaks in their study at particle diameters of ~5 µm. Additionally to fungal spores, bacteria, algae, lichens, and plant fragments, can produce mannitol and some of these can contribute to PBAP concentrations at ~5 µm. Similar assumptions are made for this study by linking FBAP to fungal spores, but chemical tracers vary between both studies. Furthermore, both literature-based emission fluxes compare local measurements to concentrations simulated on a global scale. Additional biases may arise when using theses fluxes on a regional scale.

The modeling in general is not very compelling and it seems like a missed opportunity to investigate/comment on the factors controlling the variability of PBAP. In particular, given the general skill of the model (e.g. Figure 3), it would be useful to try to separate in the model how meteorology and emissions contribute to variability (i.e. perform a simulation with constant emissions). This may be the first time that a model has been compared with high resolution PBAP observations. This is the primary unique direction of this work, but it is insufficiently explored in the current manuscript.

We will expand the discussion on the relative importance of the meteorological and emission-related variability in the manuscript. A time-independent (but spatially varying) emission flux has already been included in the simulations by $F_{S\&D}$. Three locations consist of similar surface ecosystem

properties and their emission fluxes are therefore very close to each other (F_{KA} = 782 $m^{-2}s^{-1}$; F_{Man} = 837 $m^{-2}s^{-1}$; F_{Kil} = 844 $m^{-2}s^{-1}$). The results show that diel cycles in the fungal spore concentrations also develop when using $F_{S\&D}$ for emission. In the manuscript, the description is described as follows: "By using a time-independent (but spatially varying) emission flux $F_{S\&D}$, every development in the local temporal pattern arises from meteorological influences. A similar cycle develops between constant ($F_{S\&D}$) and time-independent ($F_{H\&S}$ and F_{FBAP}) simulated fungal spore concentrations, but the order of magnitude differs to varying extend. Therefore and from by visually comparing to simulated boundary layer height, a diurnal cycle in the simulated fungal spore concentrations with a maximum between midnight and sunrise is probably influenced at least partly by boundary layer compression at night."

1. Page 9907, lines 27-30: Regarding the role of PBAP as IN, the authors may also want to comment on the work of Hoose et al, 2010 and perhaps Creamean et al., 2013 which seem quite relevant here.

Further comments on the role of PBAP acting as IN are now included and additional references are taken into account. The part has changed to:

"These bio-IN may be important for ice nucleation in mixed-phase clouds at temperatures warmer than -15°C (DeMott and Prenni, 2010). In regimes colder than that, mineral dust particles and other ice nucleators are also active and the relative atmospheric abundance of PBAP is probably too small to contribute significantly to formation and evolution of these colder clouds. Previous modelling studies suggest, that bio-IN concentrations are several orders of magnitude lower than IN concentrations from mineral dust or soot and hence the influence of bio-IN on precipitation is limited on the global scale (Hoose et al., 2010; Sesartic et al., 2012; Spracklen and Heald, 2013). *In-situ* analyses of insoluble cloud ice and precipitation residuals meanwhile highlight the contribution of bio-IN to precipitation, and back trajectories indicate that they can be transported over large distances (Creamean et al., 2013)."

2. Page 9909, lines 1 and 10 seem to contradict each other, FBAP cannot be a lower limit for PBAP if it may be contaminated by other fine particles.

Uncertainties in FBAP measurements include both, a possible underestimation of PBAP as not all PBAP cause a fluorescence signal and some smaller PBAP are excluded by size selection (<1 µm), and a possible overestimation as some non-PBAP contaminate the FBAP signal to some extent. By comparison to traditional measurement techniques for PBAP, it has been found that the former aspect dominated and that FBAP can be considered a lower limit for PBAP (Pöschl et al., 2010).

3. Section 2.1: much of this is section (page 9909, line 25 through all of page 9910) is basic model treatment of aerosols that does not need to be included in a scientific manuscript. I suggest the authors trim this (equations are not necessary).

We understand the reviewer is suggestion to shorten the methodology section, but we think that a brief description of basic aerosol treatment in the model is important, especially as we are addressing readers from both the modelling and the observational communities. Descriptions of dry and wet fungal spore deposition are especially relevant for a sufficient understanding of issues about the atmospheric lifetime of fungal spores (discussed in section 3.2).

4. Page 9911, line 14: typo "gases at particulate" should be "gases and particulate"

Corrected

5. Page 9911, line 23: what are "Anthropogenic primary aerosols"?

Aerosol emissions in PM2.5 and PM10 size mode are disaggregated into chemical components using a split table from TNO (Kuenen et al., 2011). The category "other anthropogenic primary aerosols" represents the remaining, non-carbonaceous primary part, including e.g. minerals, metal oxides, and product emissions (Knote et al., 2011).

6. Section 2.2: line 1 and line 27 incorrectly suggest that the H&S parameterization is a constant emission rate. The authors have described how it is a dynamic function of q and LAI, therefore is by definition, not constant in time or location.

Heald and Spracklen (2009) also used a constant emission rate as a first guess and corrected it afterwards in order to give a better representation of mean mannitol concentrations, which is described in eq. 6 with a function giving $F_{H\&S}$. In order to not confuse the reader in future, we decided to ignore this detail and remove the citation in the first place.

7. Page 9913, line 3: Please quantitatively compare the impact of your different size assumption (3 um) here with Hoose et al., 2010a (5um) and Heald and Spracklen, 2009 (PM2.5). Are your totals for mass, number emissions comparable? How did you scale your constant (c) term to account for the smaller size range from Hoose et al.?

A constant c = 4.6 is applied to the number emission flux of 3 µm particles in this study in order to have consistent mass emission flux with Hoose et al. (2010) and Heald and Spracklen (2009). The former is obtained by:

$$M_{this\ study} = M_{Hoose\ 2010} = \frac{\pi}{6}\ (3\ \mu m)^3\ \rho_{spore}\ \underbrace{\left(\frac{5\ \mu m}{3\ \mu m}\right)^3}_{c=4.6}\ F_{Hoose\ 2010}$$

The latter gives a maximum fungal spore emission of $M_{H\&S} \approx 1$ g/m^{2}yr in regions with highest LAI and q_v. For the same conditions of LAI and q_v, number emission flux from Hoose et al. (2010) gives $F_{H10} = 500$ m^{-2}s^{-2}, which matches $M_{H\&S}$ for $\rho_{spore} = 1$ g/cm^3 (see Figure 3a in Heald and Spracklen (2009)).

8. Page 9913, equation 6: This equation does not explicitly match the parameterization of H&S (unless one assumes that their constant=c/(LAI_max qv_max). Why is the parameterization given this way here? Is there a physical justification?

We agree that the chosen formulation including an index "max" is confusing and changed it to giving the numbers directly inside the equation.

9. Page 9913, line 11: what is the time resolution for qv (i.e. the meteorological time step)?

All meteorological parameters (including q_v) are written out hourly. When using measured FBAP concentrations for the regression analysis, they are averaged for one hour in order to be consistent to the model output.

10. Page 9913, lines 13-18: This section is confusing. There are 3 sentences that mention the IC/BC for aerosols, and it's not clear what the authors mean by "No initial and boundary concentrations are predefined for aerosols or gases." Please clarify or simplify this text.

Simplified in text:

"The COSMO-ART mesoscale model system is driven by initial and boundary data for meteorological conditions. They are updated every six hours and result from interpolation of the coarse grid operational atmospheric model analysis of the ECMWF (European Centre for Medium-Range Weather Forecasts)."

11. Figures 3-6: missing statistical quantification of model performance

See description above.

12. Figures 3-6: all 3 schemes co-vary. To what degree does variability reflect meteorology (PBL height, mixing, and precipitation) rather than variability in emissions driver (q, LAI)? This can be diagnosed in the simulation (statement on page 9919 lines 15-16 isn't quite true. Some of these effects can be deconvolved in the model). What is the correlation between FBAP and PBL height?

By comparing the simulated concentrations from both literature-based emission parameterizations ($F_{S\&D}$ = const.), influences of varying meteorological conditions and varying emission drivers are expressed. These effects cannot clearly be separated in the temporal pattern of the FBAP concentrations.

13. Page 9919, line 1: Can you show the observed precipitation from the sites in the Figures?

Exploring the correlation of FBAP concentrations to precipitation is beyond the scope of this study and is described elsewhere (e.g. Schumacher et al., 2013).

14. Page 9920, lines 7-9: Is there any evidence of this phenomenon in the observations you are exploring here?

Some of the FBAP concentrations used in this study are also included in the referenced studies (Huffman et al., 2013; Schumacher et al., 2013). An in-depth analysis of this phenomenon is beyond the scope of this study.

15. Page 9921, line 9-10: Figure 7a does not support the statement that the simulated concentrations are "systematically underestimated". Nor does Figure 7b demonstrate the improvement suggested on page 9924, line 8. Please show statistics to support these claims.

These effects are difficult to be recognize on a log-scale, but a more detailed statistical analyses is now included.

16. Page 9922, line 3-4: what is the lifetime in the model simulations?

In the model simulation, a local estimate of the fungal spore lifetime calculated as follows:

$$\tau_{spore} = \frac{\int_{z=0}^{top} N_{spore}(z)\ dz}{F_{spore}}$$

The domain-average fungal spore lifetime is calculated by:

$$\tau_{spore} = \frac{\int\int\int N_{spore}(x,y,z)\ dx\ dy\ dz}{\int\int F_{spore}(x,y)\ dx\ dy}$$

N_{spore} and F_{spore} give the fungal spore number concentration in $1/m^3$ and emissions flux in $1/m^2s$, respectively, and refer to any of the three emission parameterizations. The integral runs from surface model layer (~10 m above ground) to the top-most model layer (at a height of 24 km). The atmospheric lifetime for each time period and location as temporal average are shown in Table 2.

Table 2. Average atmospheric lifetimes of fungal spores at each location in hours.

Hyytiälä			Karlsruhe			Killarney	Manchester
July	October	August	July	October	August	August	August
1.425	1.388	0.748	3.731	6.081	2.801	1.961	1.578

These calculations reveal a strong effect of wet deposition on the simulated lifetime, which is highest for the October case in Karlsruhe (where nearly no rain occurred in the simulation) and shortest for the August case in Hyytiälä (with extensive rain in the simulation).

17. Page 9922, line 7: This lifetime is very short. What physical mechanism could justify such a rapid removal rate?

As indicated in the answers to the previous comments, the model simulates a very strong wet removal of the spores. In addition, calculating the atmospheric lifetime in this simple way assumes equilibrium between local removal and source processes. At the model boundaries, fungal spores are only removed from the domain without fungal spores being transported into the domain. This disagreement may cause an underestimation of the lifetime. Sedimentation is not a major sink for fungal spores, as the average sedimentation velocity for fungal spores is v_{sed} = 0.035 cm/s (calculated after Helbig et al., 2004), which is typical for 3 µm particles.

18. Page 9923, equation 8: Are the variables used here independent? Have you verified that you are not over-fitting?

q_v is a diagnostic variable of the model and to some extent depends on T, but an additional T-dependence has been reported for other FBAP-studies (Jones and Harrison, 2004; Di Filippo et al., 2013).

19. Figure 9: Can you overplot the observations using the same assumptions?

Figures 8 and 9 are modified such that an additional circle around each location gives the average measured FBAP concentration / FBAP emission flux colored with the same colorbar as the map.

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