Atmospheric budget of primary biological aerosol particles from fungal spores

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[1] The contribution of primary biological aerosol particles (PBAP) to the global budget of organic aerosol is poorly understood. Concentrations of mannitol, a biotracer for fungal spores, are used here to constrain the first global model (GEOS-Chem) simulation of PBAP from fungi. Emissions are driven by leaf area index and atmospheric water vapor concentrations and are empirically optimized based on the geographical and seasonal variability of observed mannitol concentrations. Optimized global emissions total 28 Tg yr^{-1}, with 25% of that total emitted as fine mode ($PM_{2.5}$) aerosol. Fungal spores contribute 23% of total primary emissions of organic aerosol, or 7% of the fine-mode source. Annual mean simulated surface concentrations of PBAP over vegetated regions range from 0.1–0.7 μgm^{-3} ($PM_{2.5}$) and 0.4–3.0 μgm^{-3} (PM_{10}), with the highest concentrations in the tropics, where PBAP may be the dominant source of organic aerosol. Further validation is required to reduce the substantial uncertainties on this budget. **Citation:** Heald, C. L., and D. V. Spracklen (2009), Atmospheric budget of primary biological aerosol particles from fungal spores, *Geophys. Res. Lett.*, *36*, L09806, doi:10.1029/2009GL037493.

[2] Primary biological aerosol particles (PBAP) may make up a large unrecognized fraction of organic aerosol in the atmosphere [*Elbert et al.*, 2007; *Jaenicke*, 2005]. A variety of cellular particles contribute to this aerosol class, including viruses, bacteria, fungal spores, pollen and vegetative debris. Certain among them play a demonstrable climate-forcing role, whether as cloud condensation nuclei (CCN) [*Bauer et al.*, 2003] or ice nuclei (IN) [*Christner et al.*, 2008; *Diehl et al.*, 2001]. However, the identification and detection of these particles is challenging and often limited to laborious staining and counting methods. *Mahowald et al.* [2008] estimated the budget of coarse bioaerosol based on a statistical attribution of the sources of phosphorous. The sugar alcohol mannitol ($C_6H_8(OH)_6$), has been identified as a unique tracer of fungal spore PBAP [*Bauer et al.*, 2008a; *Elbert et al.*, 2007]. We use observations of mannitol to develop the first global simulation of PBAP from fungal spores and examine the contribution that this source makes to the organic aerosol budget.

[3] Spores from fungi, a class of bioaerosol which can include yeasts and molds, can be sources of both infection and allergic reaction [e.g., *Green et al.*, 2003]. They are associated with vegetation, soil, litter and decaying organic matter and are emitted to the atmosphere both via active injection and passively through the action of wind which ultimately disperses spores away from sources [*Cox and Wathes*, 1995]. These spores generally span the 1–20 μm size range and have been found to dominate the coarse mode bioaerosol in certain environments [*Glikson et al.*, 1995]. Attempts have been made to quantify the contribution of fungal spores to atmospheric particulate matter based on their phospholipid [*Womiloju et al.*, 2003] or their culturable content [*DiGiorgio et al.*, 1996], but more recently sugar alcohols, such as mannitol, have been shown to be a unique biomarker for these spores [*Bauer et al.*, 2008a]. *Elbert et al.* [2007] and *Bauer et al.* [2008a] estimate that each spore contains 1.7 pg of mannitol. These studies report different values for the mean mass of organic matter associated with each spore, from 33 pg/spore [*Bauer et al.*, 2008a] to 65 pg/spore [*Elbert et al.*, 2007], however *Bauer et al.* [2002] find a wide range of carbon content for spores which may reflect variation in species of fungi. The resulting scaling from mannitol concentration to organic matter (OM) concentration may therefore range from 19 to 38 g OM/g mannitol. We use the larger value in this study and thus the fungal spore PBAP budget estimated here is likely an upper limit.

[4] *Elbert et al.* [2007] compiled measured mannitol concentrations in organic aerosol particles from diverse environments. Based on typical extratropical mannitol concentrations, they estimate a total global flux of PBAP from fungal sources of 50 Tg yr^{-1}. Emission of OC aerosol from combustion sources are estimated to total 47 Tg yr^{-1} [*Bond et al.*, 2004] (with significantly year-to-year variability) however, these particles are generally sub-micron in size. While previous studies have compared total estimated PBAP emission to these combustion sources, we attempt to assess the contribution to both the fine ($PM_{2.5}$, diameter < 2.5 μm) and the coarse (PM_{10}, diameter < 10 μm) mode here. This separation of size is critical to the interpretation of ambient organic aerosol measurements, where instrument and sampling design often dictates fine mode sampling only. In addition, this size segregation provides a crude delineation between small, longer-lived particles and larger particles which may be more rapidly deposited from the atmosphere. We use data from *Elbert et al.* [2007] as a starting point of this study and exploit the geographical and seasonal diversity of observed mannitol concentrations to constrain the emission source for both fine and coarse mode PBAP.

[5] To optimize estimates of fungal spore PBAP emissions, we implement a PBAP simulation in the GEOS-Chem global chemical transport model (http://www-as.harvard.edu/chemistry/trop/geos), driven by GEOS-4 assimilated meteorology from the NASA Global Modeling and Assim-

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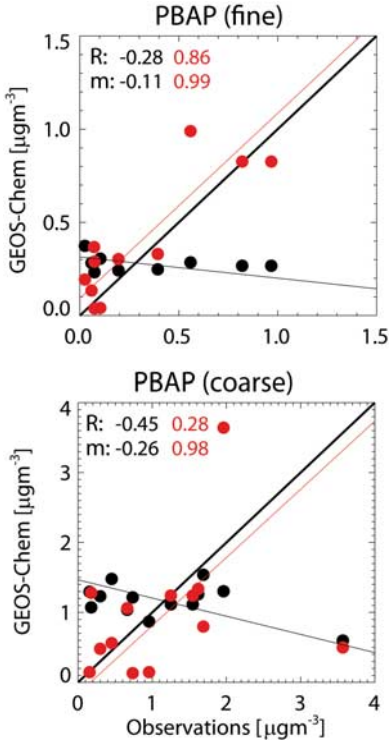

Figure 1. Point-by-point comparison between observed fungal PBAP and corresponding GEOS-Chem model simulation for both constant emissions (black) and optimized emissions (red). Observations are compared to simulated monthly or seasonal means as appropriate. Also shown are the corresponding linear regressions (solid lines) and the 1:1 relationship (thick black line). The correlation coefficient and slopes for the linear regressions are indicated. Statistics were computed with the reduced-major-axis method [*Hirsch and Gilroy*, 1984].

ilation Office. We employ here v7.04.13 of GEOS-Chem at a $2^\circ \times 2.5^\circ$ horizontal resolution for the year 2003. PBAP is added to the GEOS-Chem simulation in 2 size modes: fine (<2.5 μm) and coarse (2.5–10 μm). We assume an organic matter to carbon (OM:OC) ratio of 2.6 and thus a molecular weight of 31 g mol^{-1}[*Bauer et al.*, 2008b]. Dry deposition velocities for PBAP are calculated with a size-dependent scheme [*Zhang et al.*, 2001], matching the treatment of dust and sea salt deposition in GEOS-Chem. We assume that fungal spores can readily be integrated in cloud droplets, and are therefore treated as soluble. The implied rapid removal reduces the possible lifetime of PBAP in the atmosphere, and thus ensures that all emission estimates here are an upper-limit. Aerosol scavenging in GEOS-Chem follows the scheme of *Liu et al.* [2001]. Anthropogenic emissions of OC are as described by *Park et al.* [2003] and biomass burning emissions are from the Global Fire Emissions Database version 2 (GFEDv2) [*van der Werf et al.*, 2006]. Emissions of primary organic aerosol from these combustion sources total 66 Tg yr^{-1} in 2003. We assume that 50% of OC emitted from combustion sources is hydrophobic with a 1.2 day e-folding conversion from hydrophobic to hydrophilic [*Park et al.*, 2003]. Formation of secondary organic aerosol (SOA) from the products of isoprene, monoterpenes and other volatile organic compounds (OVOCs) oxidation follows the 2-product model as implemented by *Chung and Seinfeld* [2002] based on empirical fits to smog chamber data. Biogenic SOA production in GEOS-Chem totals 26.8 Tg yr^{-1} [*Henze et al.*, 2008]. An 80% scavenging efficiency is assumed for SOA [*Chung and Seinfeld*, 2002]. With the exception of PBAP, OC is treated as bulk submicron aerosol.

[6] We first test whether a constant emission rate can reproduce the magnitude and range of observed mannitol concentrations. These measurements reflect long-term averages ranging from weeks to months of sampling in a variety of continental locations and seasons [*Elbert et al.*, 2007, Table A3]. Measured mannitol concentrations in coarse mode particles (N = 13) exceed those in the fine mode (N = 11) by a factor of 3–4. We therefore apply the emission rate estimated by *Elbert et al.* [2007] (1.1×10^{-8} g m^{-2} s^{-1}) as 20% fine and 80% coarse mode PBAP. Global emissions total 44 Tg yr^{-1}, which is lower than the 50 Tg yr^{-1} estimated by *Elbert et al.* [2007] because we do not emit from ice and snow-covered surfaces. Figure 1 compares the resulting GEOS-Chem simulation of PBAP in the fine and coarse mode to the fungal spore PBAP observations (scaled from mannitol concentrations) in black. The GEOS-Chem simulation has little skill in capturing the variability of the observed PBAP or the range of concentrations when these constant emissions are employed. Measured mannitol concentrations reflect an important geographical variability with concentrations in Amazonia and Rondonia 2–3 times higher than those reported at extratropical locations [*Elbert et al.*, 2007]. In addition, the GEOS-Chem simulation reveals a seasonal bias, with concentrations reported in spring and winter overestimated and summer/fall concentrations underestimated. Due to the long-term sampling, the meteorological variability between particular years of observation and the 2003 simulation is not expected to be a factor in these comparisons.

[7] We attempt to optimize emissions to reflect the seasonal and geographical variability in the observations. In-situ fungal spore concentrations have been observed to vary with a range of meteorological and surface variables [*Jones and Harrison*, 2004]. We examine how observed mannitol concentrations might be related to a series of meteorological and phenological drivers including: temperature, radiation, wind speeds, surface wetness, precipitation, leaf area index (LAI), relative humidity, water vapor concentrations and boundary layer depths. Figure 2 shows the two factors most highly correlated with mannitol concentrations: LAI (a measure of vegetation density) and atmospheric water vapor concentrations. These two variables can be taken as proxies for the density of the source medium (LAI) and water availability, both of which have been seen to control long-term fungal spore concentrations [*Jones and Harrison*, 2004]. However, some of the drivers are highly correlated (for example, LAI and temperature), and so the empirical drivers shown to give the best correspondence may be surrogates for other factors controlling fungal sources. Previous studies have used annual-mean biomass density, a measure similar to LAI, to drive their total PBAP [*Mahowald et al.*, 2005, 2008]. We further note that the observations do not allow us to assess what might control the hourly-daily variability in concentrations, where factors

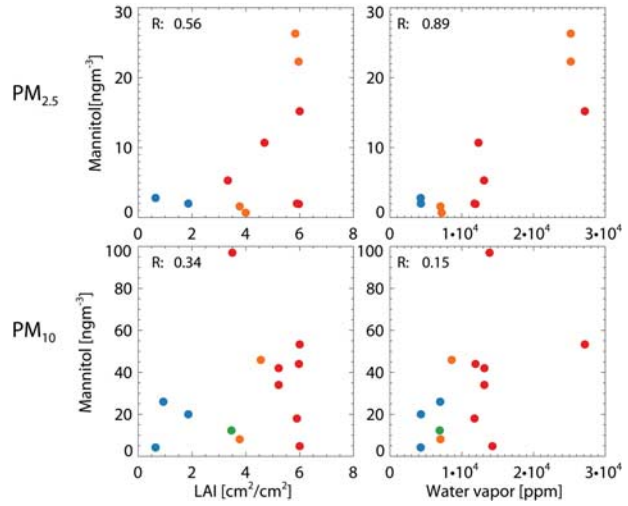

Figure 2. Correlation of observed mannitol concentrations with (left) monthly-mean LAI and (right) water vapor concentrations at each location. Points are colored by season: spring (green), summer (red), autumn (orange), and winter (blue).

such as wind speed and humidity may be more important [*Jones and Harrison*, 2004].

[8] Figure 1 compares the GEOS-Chem simulation with optimized emissions to the observations of organic aerosol from fungal spores (in red). A number of emission iterations were tested, including scaling emissions by either one of LAI or water vapor concentrations (as well as other parameters). The objective of the optimization was to minimize the mean differences between observed and simulated observations and to maximize the degree of variability reproduced by the model. Shown here are the emissions optimized to fit the mean observed concentrations, by linearly scaling a constant emission rate by both LAI and water vapor concentrations. The model captures the variability in the fine mode PBAP concentrations (R = 0.86), with little bias (slope = 0.99) when these optimized emissions are implemented. Coarse mode emissions are obtained by uniformly scaling fine mode emissions by a factor of three and thus reproduce the mean magnitude of the measured coarse concentrations (slope = 0.98). The simulation is less successful in reproducing the variability observed in the coarse mode PBAP (R = 0.28).This represents an improvement over the constant emissions scheme, although better constraints are required to establish what additional parameters might control the emission of fungal spores in the coarse mode. The global optimized emissions total 7 Tg yr^{-1} in the fine mode and 21 Tg yr^{-1} in the coarse mode. *Winiwarter et al.* [2009] used atmospheric concentrations of fungal spores observed at a site in Europe to derive a fungal spore emission estimate of 3×10^{-3} – 0.08 g m^{-2} yr^{-1}. Our emissions in the same region are higher at 0.2 g m^{-2} yr^{-1}, however if we assume the same organic matter conversion used by *Winiwarter et al.* [2009] we obtain values at the upper limit of their range. *Mahowald et al.* [2008] estimate a total coarse mode PBAP emission of 164 Tg yr^{-1} with a similar geographical distribution; taken with our results this would suggest that fungal spores make up ~12% of coarse PBAP. Fungal spores contribute 23% of total primary emissions of organic aerosol in this simulation, or 7% of the fine-mode source. Figure 3a shows the global distribution of emissions, which are highest in the tropics. In the extratropics, where vegetation cover varies significantly throughout the year, simulated emissions and concentrations exhibit a pronounced seasonal cycle, peaking

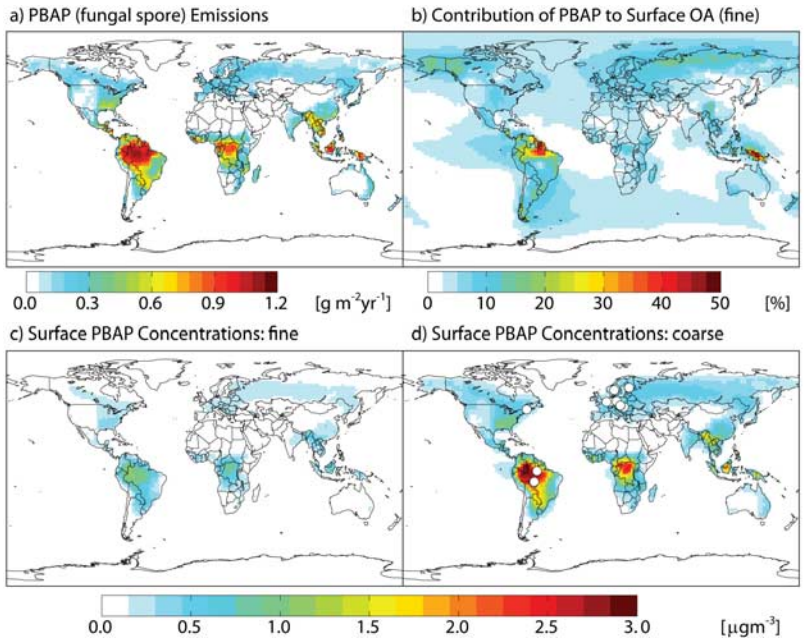

Figure 3. Annual mean of optimized GEOS-Chem simulation of fungal PBAP: (a) PBAP emissions, (b) percentage contribution of fungal PBAP to fine organic aerosol (OA) surface concentrations, (c) fine-mode fungal PBAP surface concentrations, and (d) coarse-mode fungal PBAP surface concentrations. Observation locations are shown as white circles in Figure 3d (note some locations overlap).

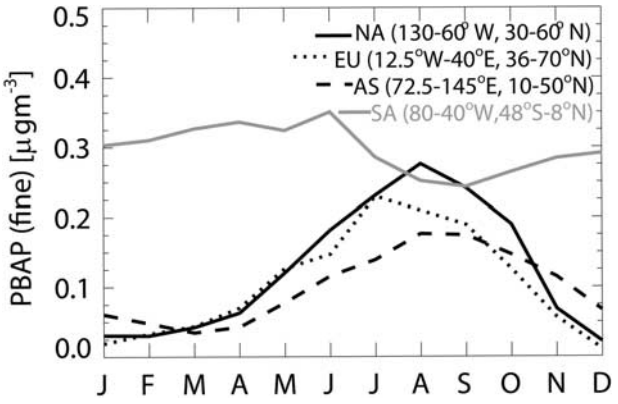

Figure 4. Seasonality in mean simulated fine mode PBAP surface concentrations over indicated domains: North America (NA, solid), Europe (EU, dotted), Asia (AS, dash) and South America (SA, grey).

in late local summer and early fall (Figure 4). Measured spore counts in urban and suburban Vienna [*Bauer et al.*, 2008b], the Hyytiala field site in Finland (H. E. Manninen, personal communication, 2009), and in Hualien, Taiwan [*Ho et al.*, 2005] confirm this seasonality.

[9] Figure 3 also shows annual mean simulated surface concentrations in both the fine and coarse mode. Concentrations of fine-mode PBAP rarely exceed 0.3 μg m^{-3}, while concentration of coarse-mode PBAP are typically 0.4 μg m^{-3} or larger over vegetated regions. If we convert typical concentrations of 0.1 μg m^{-3} and 0.5 μg m^{-3} for the fine and coarse mode respectively to equivalent number concentrations (assuming a density of 1 g cm^{-3} and diameters in the middle of each size range) we obtain number concentrations of 10^5 m^{-3} (fine) and 8×10^3 m^{-3} (coarse). *Elbert et al.* [2007] suggest that spore concentrations are typically less than 10^4 m^{-3} but can reach up to 10^6 m^{-3}. Our values are very sensitive to the assumed spore diameter. For example, assuming that the mean diameter of the fine mode aerosol is 2 μm rather than 1.25 μm would reduce the fine mode number concentration by a factor of 4. In a future study, which will focus on the contribution of these particles to CCN activity, we will investigate how the size of these particles can be optimized to match fungal spore counts, while matching mass emissions estimated here.

[10] The simulated annual mean burden of PBAP from fungal sources determined from GEOS-Chem and the optimized emission model is 0.18 Tg. Fine mode PBAP makes up a larger fraction of the burden (30%) than its proportional emission (25%) due to the longer lifetime of the smaller size mode (global mean lifetime of 2.8 days for fine mode and 2.2 days for coarse mode). Figure 3b shows that the percentage of fine mode organic aerosol at the surface over land that can be attributed to PBAP from fungal sources is of the order of 10% but can exceed 30% in the tropics. In the free troposphere concentrations of fungal spores are less than 0.05 μg m^{-3} and contribute less than 5% to fine organic aerosol. Assumptions in the model were specified consistently such that these values should be taken as an upper-estimate.

[11] PBAP from fungal sources represent an important addition to the global organic aerosol budget, particularly over clean, densely vegetated environments. Uncertainties on the fungal spore PBAP budget estimated here are large, particularly given the limited set of observations used to optimize emissions. There is a critical need for more extensive datasets against which this empirical model simulation can be evaluated. This includes measurements in diverse environments, over multiple seasons with high temporal sampling. Much of this aerosol may fall within the coarse size range, motivating a need for further size-resolved field observations. The contribution of this fungal source to CCN (and IN) concentrations now needs to be evaluated as do implications for climate and the potential for PBAP-climate feedbacks. Field observations suggest that primary biological aerosol particles are ubiquitous [*Cox and Wathes*, 1995]. This study represents a first-step towards building a global atmospheric source-specific bioaerosol budget.

[12] **Acknowledgment.** We thank Tony Prenni for useful discussions.

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