

1 Analysis of particulate emissions from tropical biomass 2 burning using a global aerosol model and long-term 3 surface observations

4 C.L.Reddington¹, D.V. Spracklen¹, P. Artaxo², D.A. Ridley^{1,3}, L.V. Rizzo⁴ and A.
5 Arana²

6 [1] {School of Earth and Environment, University of Leeds, Leeds, United Kingdom}

7 [2] {Department of Applied Physics, Institute of Physics, University of Sao Paulo, Sao Paulo,
8 Brazil}

9 [3] {now at Department of Civil and Environmental Engineering, Massachusetts Institute of
10 Technology, USA}

11 [4] {Institute of Environmental, Chemical and Pharmaceutical Sciences, Federal University of
12 Sao Paulo, Diadema, Brazil}

13 Correspondence to: C. L. Reddington (c.l.s.reddington@leeds.ac.uk)

14 15 Abstract

16 We use the GLOMAP global aerosol model evaluated against observations of surface
17 particulate matter (PM_{2.5}) and aerosol optical depth (AOD) to better understand the impacts of
18 biomass burning on tropical aerosol over the period 2003 to 2011. Previous studies report a
19 large underestimation of AOD over regions impacted by tropical biomass burning, scaling
20 particulate emissions from fire by up to a factor 6 to enable the models to simulate observed
21 AOD. To explore the uncertainty in emissions we use three satellite-derived fire emission
22 datasets (GFED3, GFAS1 and FINN1) in the model. In these emission datasets the tropics
23 accounts for 66-84% of global particulate emissions from fire. With all emission datasets the
24 model underestimates dry season PM_{2.5} concentrations in regions of high fire activity in South
25 America and underestimates AOD over South America, Africa and Southeast Asia. **When we**
26 **assume an upper estimate of aerosol hygroscopicity**, underestimation of AOD over tropical
27 regions impacted by biomass burning is slightly reduced, relative to previous studies. Where
28 coincident observations of surface PM_{2.5} and AOD are available we find a greater model
29 underestimation of AOD than PM_{2.5}, **even when we assume upper estimates of aerosol**
30 **hygroscopicity**. Increasing particulate emissions to improve simulation of AOD can therefore

1 lead to overestimation of surface PM_{2.5} concentrations. With FINN1 emissions scaled by a
2 factor of 1.5 the model reasonably simulates PM_{2.5} concentrations and AOD in South America
3 and AOD over Southeast Asia, but underestimates AOD over Africa. GFAS1 emissions require
4 a scaling of 3.4 for the model to match observed PM_{2.5} and AOD, with the exception of
5 Equatorial Asia where a scaling factor of 1.5 is adequate. The model with GFED3 emissions
6 scaled by a factor of 1.5 reasonably simulates PM_{2.5} concentrations and AOD in active
7 deforestation regions in South America and AOD in Equatorial Asia, but requires a larger
8 scaling factor to capture observed AOD in Africa, Indochina and elsewhere in South America.
9 The model with GFED3 emissions poorly simulates observed seasonal variability of surface
10 PM_{2.5} and AOD in regions where small fires dominate, providing independent evidence that
11 GFED3 omits emissions from small fires. Seasonal variability of both PM_{2.5} and AOD in South
12 America is better simulated by the model using FINN1 and GFAS1 emissions. Detailed
13 observations of aerosol properties over biomass burning regions are required to better constrain
14 particulate emissions.

15

16 **1. Introduction**

17 Open biomass burning is an important source of trace gases and particulate matter (PM) to the
18 atmosphere (Crutzen and Andreae, 1990; Andreae and Merlet, 2001; Van der Werf et al., 2010).
19 Biomass burning emissions can influence weather (Kolusu et al., 2015; Gonçalves et al., 2015;
20 Tosca et al., 2015) and climate (Ramanathan et al., 2001; Tosca et al., 2013; Jacobson, 2014)
21 directly, by scattering and absorbing solar radiation (Johnson et al., 2008; Sakaeda et al., 2011),
22 and indirectly, by modifying cloud properties (Andreae et al., 2004; Feingold et al., 2005; Tosca
23 et al., 2014). The influence of biomass burning aerosol on surface radiation can have subsequent
24 impacts on the biosphere. For example, smoke plumes from biomass burning have been
25 observed to increase plant productivity, through increasing the amount of diffuse radiation
26 (Oliveira et al., 2007; Doughty et al., 2010), which has been shown to be a regionally important
27 process over the Amazon (Rap et al., 2015). PM from biomass burning can substantially
28 degrade regional air quality leading to adverse effects on human health (Emmanuel, 2000;
29 Frankenberg et al., 2005; Johnston et al., 2012; Jacobson, 2014; Reddington et al., 2015). A
30 better understanding of particulate emissions is needed to improve predictions of the impacts
31 on biomass burning on climate and air quality. Here we use a global aerosol model with tropical
32 observations of surface PM and aerosol optical depth (AOD) to better understand the impact of
33 tropical fires on atmospheric aerosol.

1 The spatial and temporal distribution of fires depends on climate, vegetation and human
2 activities. At the global scale, fire emissions are dominated by burning in the tropics (van der
3 Werf et al., 2010). Anthropogenic activity can increase the occurrence of fires either directly,
4 through deforestation fires and agricultural residue burning (van der Werf et al., 2010), or
5 indirectly, through land-use/land-cover change that acts to increase the fire susceptibility of the
6 land surface e.g. forest fragmentation in the Amazon (Cochrane and Laurance, 2002) and large-
7 scale drainage of peatlands in Indonesia (Field et al., 2009; Carlson et al., 2012). Human activity
8 can also reduce the occurrence of fires, directly through fire suppression and indirectly through
9 reducing and fragmenting fuel loads which limits fire spread (Bistinas et al., 2014). Over the
10 21st century, predicted changes in rainfall and temperature may increase forest water stress and
11 subsequent fire occurrence in tropical forests (Cox et al., 2008; Golding and Betts, 2008; Malhi
12 et al., 2009). The incidence of fire and resulting emissions are therefore sensitive both to
13 changing climate and changes in land-use (Heald and Spracklen, 2015).

14 High temporal and spatial variability in biomass burning emissions coupled with the difficulties
15 involved in conducting measurements in remote tropical regions lead to major challenges for
16 their quantification. In recent years, global estimates of biomass burning emission fluxes have
17 mostly been obtained using satellite remote sensing (e.g., van der Werf et al., 2006, 2010; Reid
18 et al., 2009; Wiedinmyer et al., 2011; Kaiser et al., 2012; Zhang et al., 2012; Ichoku and Ellison,
19 2014), which provides long-term observations with relatively high spatial coverage. A range of
20 satellite products and methods are utilised to derive fluxes of aerosol and gas-phase species
21 emitted from fires. The most common methods use satellite-retrieved burned area, active fire
22 counts, and/or fire radiative power (FRP) in combination with biogeochemical models (when
23 using burned area) and/or species-specific emission factors obtained from laboratory
24 experiments and field observations (e.g., Hoelzemann et al., 2004; Ito and Penner, 2004; 2005;
25 van der Werf et al., 2006, 2010; Wiedinmyer et al., 2006; 2011; Schultz et al., 2008; Kaiser et
26 al., 2012). Large uncertainties are associated with satellite observations of fires and with the
27 various methods used to calculate emissions fluxes from the observational data (e.g. Ito and
28 Penner, 2005; Reid et al., 2009; Konovalov et al., 2014)

29 Previous studies using satellite-derived emissions and atmospheric models to investigate the
30 properties and impacts of biomass burning aerosol have found a persistent underestimation of
31 AOD observed in most tropical biomass burning regions (Matichuk et al., 2007; 2008; Chin et
32 al., 2009; Petrenko et al., 2012; Kaiser et al., 2012; Ward et al., 2012; Tosca et al, 2013; Pereira
33 et al., 2016). In general, modelling studies have required biomass burning emissions or

1 concentrations of biomass burning aerosol to be increased by factors ranging from ~1.5 to ~6
2 in order to match satellite and ground based observations of AOD (Matichuk et al., 2007; 2008;
3 Johnson et al., 2008; Sakaeda et al., 2011; Johnston et al., 2012; Kaiser et al., 2012; Tosca et
4 al., 2013; Marlier et al., 2013). The underestimation of AOD observed in biomass burning
5 regions has been attributed to a number of factors (see e.g., Kaiser et al., 2012) including: i)
6 underestimation of biomass burning emission fluxes; ii) errors in modelling the atmospheric
7 distribution and properties of biomass burning aerosol; and iii) uncertainties in the calculation
8 of AOD.

9 Uncertainties associated with the derivation of emission fluxes arise from errors present in the
10 satellite-detection of active fires or burned area (e.g. obscuring of the surface by clouds and
11 smoke, satellite spatial resolution and detection limits, and satellite overpass time), as well as
12 uncertainties in emission factors and fuel consumption estimates. For example, Randerson et
13 al. (2012) suggest that emission datasets based on relatively coarse burned area data (detection
14 limit of ~100 Ha), result in an underestimation of global area burned by ~35%, although this
15 error is not sufficient to fully explain the underestimation of AOD discussed above. Inadequate
16 representation of biomass burning aerosol in models, including errors in the modelled aerosol
17 size distribution, chemical composition, ageing processes, vertical and horizontal transport
18 (including fire emission injection heights) and dry/wet removal from the atmosphere, could also
19 contribute to an underestimation of AOD. The contribution of secondary organic aerosol (SOA)
20 from the oxidation of volatile organic compounds in biomass burning plumes is also a large
21 uncertainty (Jathar et al., 2014; Shrivastava et al., 2015). In the calculation of AOD itself, the
22 uncertainties associated with the assumed optical properties of biomass burning aerosol e.g.
23 their refractive indices, hygroscopicity (uptake of water onto the aerosol), and/or mixing state
24 (i.e. treated as core/shell mixtures, internally/externally mixed etc.) may also contribute to this
25 negative model bias in AOD.

26 Using only AOD to evaluate estimates of biomass burning aerosol emissions can be misleading
27 because AOD depends on many factors in addition to aerosol abundance. Scaling biomass
28 burning emissions to match observed AOD could therefore lead to inaccurate model
29 representation of biomass burning aerosol concentrations and, subsequently, errors in model
30 predictions of the air quality and climate effects of biomass burning aerosol. Although there
31 has been extensive use of AOD retrievals to evaluate model predictions of biomass burning
32 aerosol, thus far there have been relatively few studies to use aerosol measurements to
33 thoroughly evaluate these models (e.g., Liousse et al., 2010; Daskalakis et al., 2015).

1 In this study, we evaluate a global aerosol microphysics model against observations of aerosol
2 mass concentrations in addition to AOD to better understand the discrepancy in modelled
3 biomass burning AOD and to ultimately improve estimates of biomass burning aerosol. We
4 also compare three different biomass burning emission inventories to investigate regional
5 differences between emissions and identify the best fit emissions for future modelling studies.
6

7 **2. Observations**

8 To evaluate the simulated distribution of PM at the surface, we use long-term *in-situ*
9 measurements of PM_{2.5} (particulates with aerodynamic diameters < 2.5 μm) mass
10 concentrations conducted at four ground stations in the Amazon region (Alta Floresta, Porto
11 Velho, Santarem and Manaus). The location and observation period are detailed for each station
12 in Table S1 in the supplementary material. Figure S1 shows the measured PM_{2.5} concentrations
13 at each station between 2003 and 2011, demonstrating the data coverage.

14 The PM_{2.5} measurements were made using gravimetric filter analysis and the measurement
15 duration ranges from less than 1 day to more than 10 days. Particles were sampled under
16 ambient relative humidity (RH) conditions (typically in the range of 80-100% RH). The
17 sampled filters were weighed after 24 hours of equilibration at 50% RH and 20°C. Amazonian
18 submicrometer aerosol particles have growth factors of ~1.1-1.3 at 90% RH (Zhou et al, 2002;
19 Rissler et al., 2006) so we estimate that water represents roughly ~10-20% of the PM_{2.5} mass
20 concentrations at measurement conditions. Uncertainties related to filter handling, sampling
21 and analysis are estimated as 15% of particle mass. Further information on the measurements
22 conducted at the Manaus and Porto Velho stations can be found in Artaxo et al. (2013). Our
23 evaluation of PM_{2.5} is restricted to Amazonia since there are few long-term observations of
24 PM_{2.5} in other tropical regions impacted by biomass burning.

25 The measurement stations at Porto Velho and Alta Floresta are located in the arc of
26 deforestation and are strongly impacted by fresh biomass burning emissions (Fig. 1). The
27 Santarem and Manaus stations are located within forest reservations and are impacted by
28 transported regional biomass burning emissions in the dry season. The Santarem station is
29 located in Para, where the number of fire hotspots observed by satellites during the dry season
30 are typically a factor of ~10 greater than the number observed in Amazonas, where the Manaus
31 station is located. Thus in the dry season, PM_{2.5} concentrations measured at Santarem are
32 typically higher than those measured at Manaus.

1 To evaluate the simulated distribution of AOD, we use observations of spectral columnar AOD
2 measured by the Aerosol Robotic Network (AERONET) using ground-based Cimel sun
3 photometers (Holben et al., 1998). Specifically, we use Level 2.0 (quality assured) daily average
4 AOD retrieved at 440 nm from 27 AERONET stations detailed in Table S1. We selected
5 stations located within regions influenced by tropical biomass burning (Southeast and
6 Equatorial Asia, Central and Southern Africa, and the Amazon region in South America) that
7 have more than one year of relatively continuous data (automatic cloud screening leads to gaps
8 in the dataset) between 2003 and 2011. We note that whilst the majority of cloud-contaminated
9 AOD data is removed; comparisons with co-located Micro-Pulse Lidar Network observations
10 indicate that some contamination from thin cirrus clouds may remain, possibly leading to small
11 positive biases in observed AOD (Huang et al., 2011; Chew et al., 2011).

12 To compare modelled and observed PM_{2.5} and AOD, daily-mean model output was linearly
13 interpolated to the location (latitude, longitude and altitude above sea level) of each ground
14 station. Model data that corresponded to gaps in the observation datasets were removed prior
15 to calculating monthly-mean values used in the analysis. The modelled PM_{2.5} concentration is
16 calculated for dry aerosol, omitting the contribution of water to the total mass, thus modelled
17 PM_{2.5} concentrations may be underestimated compared to the observations, which include
18 some contribution from the mass of water.

19

20 **3. Model description**

21 **3.1 Global aerosol microphysics model**

22 The global distribution of aerosol was simulated using the 3-D Global Model of Aerosol
23 Processes (GLOMAP; Spracklen et al., 2005a,b; Mann et al., 2010), which is an extension to
24 the TOMCAT chemical transport model (Chipperfield, 2006). Simulations were run for the
25 period 2003 to 2011. Large scale atmospheric transport and meteorology in TOMCAT are
26 specified from European Centre for Medium-Range Weather Forecasts (ECMWF) analyses,
27 updated every 6 hours and linearly interpolated onto the model time-step. The model runs at a
28 horizontal resolution of $2.8^{\circ} \times 2.8^{\circ}$ with 31 vertical model levels between the surface and 10 hPa.
29 The vertical resolution in the boundary layer ranges from ~60 m near the surface to ~400 m at
30 ~2 km above the surface. GLOMAP has been extensively evaluated in previous studies against
31 aerosol observations (Mann et al., 2010, 2014; Spracklen et al., 2011a,b; Schmidt et al., 2012;
32 Scott et al., 2014; Reddington et al., 2011, 2013, 2014). Below we describe the features of the

1 model relevant for this study, please see Spracklen et al. (2005a) and Mann et al. (2010) for
2 more detailed descriptions of the model.

3 GLOMAP simulates the mass and number of size resolved aerosol particles in the atmosphere,
4 including the influence of aerosol microphysical processes on the particle size distribution.
5 These processes include nucleation, coagulation, condensation, ageing, hygroscopic growth,
6 cloud processing, dry deposition, and nucleation/impact scavenging. The aerosol particle size
7 distribution is represented using a two-moment modal scheme with seven log-normal modes
8 (Mann et al., 2010). Within each mode, aerosol particles are treated as internally mixed.
9 GLOMAP treats the following aerosol species: black carbon (BC), particulate organic matter
10 (POM), sulphate (SO₄), sea spray and mineral dust. Biogenic SOA is formed in the model via
11 the reaction of biogenic monoterpenes with O₃, OH and NO₃, which produces a gas-phase
12 oxidation product that condenses with zero vapour pressure onto pre-existing aerosol
13 (Spracklen et al., 2006, 2008). Concentrations of oxidants are specified using monthly-mean 3-
14 D fields at 6-hourly intervals from a TOMCAT simulation with detailed tropospheric chemistry
15 (Arnold et al., 2005) linearly interpolated onto the model time-step. Monthly mean emissions
16 of biogenic monoterpenes are taken from the Global Emissions Initiative (GEIA) database
17 (Guenther et al., 1995). Size-resolved emissions of mineral dust are prescribed from daily-
18 varying emissions fluxes provided for AEROCOM (Dentener et al., 2006).

19 For this study, anthropogenic emissions of sulphur dioxide (SO₂), BC and organic carbon (OC)
20 were specified using the MACCity emissions inventory (Lamarque et al., 2010; Granier et al.,
21 2011), which provides annually varying emissions for the period 1979-2010. For simulations
22 in the year 2011 we used MACCity anthropogenic emissions from 2010. Biomass burning
23 emissions of SO₂, BC and OC were specified using three different satellite-derived emission
24 datasets, which are described in detail in Section 3.3. We convert OC to POM using a prescribed
25 POM:OC ratio of 1.4, which is at the lower end of the range prescribed in other global models
26 (1.4 to 2.6) (Tsigaridis et al., 2014). The fire emissions were injected into the model over six
27 ecosystem-dependent altitudes between the surface and 6 km recommended by Dentener et al.
28 (2006). In the regions studied in this paper (South America, Africa and Southeast Asia), the fire
29 emission injection heights range between the surface and an altitude of ~3 km asl. The largest
30 fraction of the fire emissions, ranging from ~99% of emissions in Equatorial Asia to 88% in
31 Indochina, are injected below 1 km asl (or at surface level if the altitude of the model level
32 exceeds 1 km asl). Analysis of smoke plume heights has demonstrated that most smoke
33 emissions from fires occur within the boundary layer (Val Martin et al., 2010).

1 Primary carbonaceous aerosol particles are assumed to be non-volatile and are emitted into the
2 model with a fixed log-normal size distribution, assuming a number median diameter of 150
3 nm for biomass burning emissions and 60 nm for fossil fuel emissions and modal width (σ) of
4 1.59. Several previous studies have investigated the impacts of the uncertainty in the assumed
5 emission size distribution on simulated aerosol and cloud condensation nuclei concentrations
6 (Pierce et al., 2007; Pierce and Adams, 2009; Reddington et al., 2011; 2013; Lee et al., 2013)
7 and aerosol radiative forcing (Bauer et al., 2010; Spracklen et al., 2011b; Carslaw et al., 2013).
8 An assumption of a number median diameter of 150 nm for biomass burning emissions is
9 reasonably consistent with measurements of the size distributions of fresh biomass burning
10 aerosol from grassland (100 – 125 nm) and deforestation (100 – 130 nm) fires (Reid et al., 2005
11 and references therein). Once emitted into the model, the components of primary carbonaceous
12 aerosol (BC and OC) are assumed to mix instantaneously and are initially treated as non-
13 hygroscopic. Once these particles have accumulated 10 monolayers of soluble material
14 (assumed to be SOA and H₂SO₄) through condensation, they are transferred directly to the
15 corresponding soluble Aitken or accumulation mode to account for ageing. For a discussion of
16 the treatment of organic aerosol within global aerosol models see Tsigaridis et al. (2014).

17 **3.2 Calculation of aerosol optical depth**

18 AOD was calculated from the simulated aerosol size distribution using Mie theory assuming
19 spherical particles (Grainger et al., 2004) that are externally mixed within each log-normal
20 mode. For this study, modelled AOD was calculated at a wavelength of 440 nm using
21 component-specific refractive indices at the closest wavelength available (468 nm) from
22 Bellouin et al. (2011). Water uptake plays a significant role in determining AOD, altering the
23 refractive index and the size distribution of the aerosol. The water uptake for each soluble
24 aerosol component is calculated on-line in the model according to Zdanovskii-Stokes-Robinson
25 (ZSR) theory, which estimates the liquid water content as a function of solute molarity (Stokes
26 and Robinson, 1966). For POM in the soluble modes, we assign a hygroscopicity consistent
27 with a water uptake per mole at 65% of that of SO₄ (Mann et al., 2010). This is an upper estimate
28 of aerosol hygroscopicity. In section 4.1.3 we explore the sensitivity of simulated AOD to
29 different assumptions on aerosol hygroscopicity as well as aerosol refractive indices and aerosol
30 mixing state. The resulting daily-mean wet radii and refractive indices are used to calculate the
31 daily-mean aerosol extinction. Using hourly-mean values of water uptake increased simulated
32 daily AOD on average by less than 1%.

1 **3.3 Biomass burning emissions**

2 In this study we compare three different satellite-derived datasets of biomass burning
3 emissions: the Global Fire Emissions Database version 3 (GFED3; van der Werf et al., 2010),
4 the National Centre for Atmospheric Research Fire Inventory version 1.0 (FINN1; Wiedinmyer
5 et al., 2011) and the Global Fire Assimilation System version 1.0 (GFAS1; Kaiser et al., 2012).
6 The key aspects of these emission inventories are summarised in Table 1. We complete
7 GLOMAP simulations for the period 2003 to 2011 where all three emission datasets are
8 available.

9 GFED3 provides monthly-mean fire emissions of aerosol and gas-phase species from 1997 to
10 2011 at $0.5^\circ \times 0.5^\circ$ resolution (van der Werf et al., 2010). GFED3 emissions are derived using
11 the monthly-mean time series of global burned area estimates from Giglio et al. (2010). For
12 1997-2000, the fire emissions are based on burned area derived from the TRMM Visible and
13 Infrared Scanner (VIRS) and Along-Track Scanning Radiometer (ATSR) active fire data and
14 estimates of plant productivity derived from observations from the Advanced Very High
15 Resolution Radiometer (AVHRR). For November 2000 onwards, the fire emissions are based
16 on estimates of burned area, active fire detections, and plant productivity from the MODerate
17 resolution Imaging Spectroradiometer (MODIS) instrument on-board the Terra and Aqua
18 satellites. To derive total carbon emissions the satellite datasets are combined with estimates of
19 fuel loads and combustion completeness for each monthly time step from the Carnegie-Ames-
20 Stanford-Approach biogeochemical model. The carbon emission fluxes are converted to trace
21 gas and aerosol emissions using species specific emission factors compiled by Andreae and
22 Merlet (2001). From 2003 onwards, GFED3 fire emissions are available on a daily time step,
23 developed using detections of active fires from MODIS (Mu et al., 2011). Daily GFED3 fire
24 emissions were implemented in GLOMAP for the period 2003-2011.

25 FINN1 provides daily fire emissions of aerosol and gas-phase species from 2002 to 2012 on a
26 1 km^2 grid (Wiedinmyer et al., 2011). FINN1 fire emissions are based on detections of active
27 fires (specifically their location and timing) from the MODIS Fire and Thermal Anomalies
28 Product (Giglio et al., 2003). FINN1 also uses the MODIS Land Cover Type product to specify
29 land cover classes and the MODIS Vegetation Continuous Fields product to identify the
30 fractions of tree and non-tree vegetation, and bare ground. Specifically, the emitted mass (E) of
31 a certain species (i) is calculated using the following equation (Seiler and Crutzen, 1980):

$$32 \quad E_i = A(x, t) \times B(x) \times FB \times ef_i \quad (1)$$

1 Where A is the area burned at time t and location x , B is the biomass loading at location x , FB
2 is the fraction of that biomass that is burned and ef is the emission factor of species i . For each
3 fire count the area burned, A , is assumed to be 0.75 km^2 for fires detected on grassland and
4 savannah land cover classes, and 1 km^2 for those detected on all other land cover classes
5 following Wiedinmyer et al. (2006) and Al-Saadi et al. (2008). Adjustments are made to the
6 assumed burned area if the fire pixel extends partially over bare ground (reducing the burned
7 area by the percentage of bare area assigned to that pixel). Estimates of biomass loading, B , are
8 taken from Hoelzemann et al. (2004) and are assumed to be land cover specific. The fraction of
9 biomass assumed to burn, FB , in each fire pixel is determined as a function of tree cover using
10 relationships from Ito and Penner (2004) (see Wiedinmyer et al., 2006). Emission factors, ef ,
11 for each species are taken from Akagi et al. (2011).

12 GFAS1 provides daily fire emissions of aerosol and gas-phase species from March 2000 to
13 2013 at $0.5^\circ \times 0.5^\circ$ resolution (Kaiser et al., 2012). Like FINN1, GFAS1 uses the observed geo-
14 location of active fires from the MODIS instrument. However, GFAS1 also makes use of the
15 NASA fire products (MOD14 and MYD14) that provide quantitative information on the
16 radiative power of detected fires (Justice et al., 2002; Giglio, 2005). The FRP fields are
17 corrected for observation gaps due to partial cloud-cover by assuming the same FRP areal
18 density throughout the grid cell. Data assimilation is used to further fill observation gaps using
19 information from earlier FRP observations (see Kaiser et al., 2012). Spurious signals from
20 volcanoes, gas flares and other industrial activity are removed from the data. The FRP is
21 converted to the combustion rate of dry matter using land-cover-specific conversion factors
22 based on data from GFED3 (Heil et al., 2010; Kaiser et al., 2012). As for GFED3, species
23 emission rates are calculated using updated emission factors based on Andreae and Merlet
24 (2001).

25 Table 1 gives the total annual amounts of BC and OC aerosol emitted from fires over the tropics
26 for each emission inventory. The total BC and OC emitted from fires in the tropics make up 77-
27 84% and 66-77%, respectively of the global total emissions. FINN1 has the greatest tropical
28 OC emission, with emissions being 47% greater than in GFAS1 and 30% greater than GFED3.
29 Emission of BC is more consistent, with FINN1 BC emissions being 13% greater than GFAS1
30 and 1% greater than GFED3. This results in different OC:BC emission ratios between the
31 datasets with the mean ratio across the tropics varying from 10.0 in FINN1, 7.9 in GFED3 and
32 7.1 in GFAS1.

1 Figure 1a-c shows the spatial distribution of annual total biomass burning emissions of OC from
2 each fire inventory averaged over the period of 2003 to 2011. There are similarities in the
3 general spatial distributions of fire emissions, with all three inventories showing maximum
4 emissions over the tropical savannah and humid subtropical regions of Africa, the arc of
5 deforestation in Amazonia, coastal regions of Indonesia (Sumatra and Kalimantan), northern
6 Australia, and parts of Indochina (particularly Cambodia, Laos and Myanmar). However, Figs.
7 1d-f show that there are strong regional differences between the different emission inventories.
8 Differences between FINN1 and GFAS1 (Fig. 1e) and FINN1 and GFED3 (Fig. 1f) are more
9 spatially organised than differences between GFAS1 and GFED3 (Fig. 1d), which are more
10 spatially heterogeneous.

11 Over Africa, GFED3 gives higher OC emissions in northern tropical savannah and southern
12 humid subtropical regions, with GFAS1 and FINN1 giving higher emissions than GFED3 at
13 the boundaries of these regions and over central Africa. Over Australia, GFED3 gives the
14 highest OC emissions estimates over the tropical savannah region of northern Australia, with
15 GFAS1 giving the highest emissions in the dryer grassland and desert regions further south.

16 Over South America the picture is more complex. In general, FINN1 and GFAS1 emission
17 estimates are higher in northern and eastern Brazil than GFED3, with GFAS1 giving the highest
18 emissions over eastern areas and FINN1 over northern Brazil. FINN1 emissions are generally
19 higher than GFAS1 and GFED3 over the central and southern Amazon region (particularly over
20 the state of Mato Grosso), Peru and generally over northern South America. GFED3 emissions
21 are higher than FINN1 and GFAS1 in northern parts of Bolivia and the northern part of the state
22 of Rondônia in the arc of deforestation.

23 Over South Asia, Indochina and Equatorial Asia, FINN1 gives higher emissions than both
24 GFED3 and GFAS1, particularly over Bangladesh, Myanmar and Laos, with the exception of
25 the coastal peatland regions of Sumatra and Kalimantan where GFAS1 and GFED3 give higher
26 emissions than FINN1. The differences in emissions over Indonesia may be explained by a
27 potentially improved representation of tropical peat fire emissions in GFED3 and GFAS1
28 relative to FINN1 (Andela et al., 2013).

29

1 4. Results

2 4.1 Overview of all comparisons

3 4.1.1 Particulate matter concentrations in the Amazon region

4 Figure 2 shows simulated versus observed multi-annual monthly mean PM2.5 concentrations
5 at each of the four ground stations in the Amazon region (see Fig. 1 for site locations). To
6 quantify the agreement between model and observations, we use the Pearson correlation
7 coefficient (r) and normalised mean bias factor (NMBF) as defined by Yu et al. (2006):

$$8 \quad NMBF = \frac{(\sum M_i - \sum O_i)}{|\sum M_i - \sum O_i|} \left[\exp \left(\left| \ln \frac{\sum M_i}{\sum O_i} \right| \right) - 1 \right]$$

9 where M and O represent the multi-annual monthly mean model and observed values,
10 respectively, for each month i . A positive NMBF indicates the model overestimates the
11 observations by a factor of NMBF+1. A negative NMBF indicates the model underestimates
12 the observations by a factor of 1-NMBF.

13 Figure 2 demonstrates the important contribution of biomass burning to PM2.5 concentrations
14 across the region: there is a strong improvement in the agreement between model and
15 observations when biomass burning emissions are included in the model (Fig. 2b-d; NMBF =
16 0.62 to -0.25, $r^2=0.77-0.83$) relative to the simulation without fire emissions (Fig. 2a; NMBF=
17 -1.85, $r^2=0.44$).

18 The overall bias between model and observations is smallest with FINN1 emissions (NMBF=
19 -0.25) compared to GFED3 (NMBF= -0.49) or GFAS1 (NMBF= -0.62), with simulated
20 monthly mean concentrations mostly within a factor of ~2 of the observations. The correlation
21 between model and observations across all sites is relatively similar between the three emission
22 datasets, with a slightly stronger correlation with GFED3 emissions ($r^2=0.83$) compared to
23 FINN1 ($r^2=0.77$) and GFAS1 ($r^2=0.79$).

24 The NMBF and correlation between model and observations are shown for the individual
25 stations in Fig. 3a. Correlations are calculated between simulated and observed multi-annual
26 monthly mean concentrations to evaluate the ability of the model to simulate seasonal
27 variability in aerosol. In general, the model with fire emissions overestimates observed PM2.5
28 concentrations at the forest site near Manaus (mean NMBF=0.57) but underestimates observed
29 PM2.5 concentrations at the sites that are more strongly impacted by biomass burning (Porto

1 Velho, Alta Floresta and Santarem; mean NMBF= -0.60). Figure 3 demonstrates that the
2 relatively small bias with the FINN1 emissions in Fig. 2 is partly due to an overestimation of
3 PM_{2.5} concentrations at Manaus (NMBF=0.98), but also due to smaller model biases at the
4 three other sites (-0.51 to -0.11) compared to GFED3 (-0.76 to -0.48) and GFAS1 (-1.26 to -
5 0.39).

6 Figure 4 shows the multi-annual average seasonal cycle in observed and simulated PM_{2.5}
7 concentrations at the four measurement sites (the full time-series at each site is shown in Fig.
8 S1 in the supplementary material). The model with biomass burning emissions simulates the
9 observed seasonal variability in PM_{2.5} concentrations over the Amazon region, characterised
10 by high concentrations in the local dry season (between ~June to ~December depending on the
11 site) and relatively low concentrations in the wet season. At Porto Velho, Santarem and Alta
12 Floresta, the model underestimates observed PM_{2.5} concentrations during the dry season and
13 has relatively good agreement during the wet season. This suggests that the negative model bias
14 in the dry season is largely due to uncertainty in the biomass burning emissions rather than
15 anthropogenic emissions, biogenic SOA or microphysical processes in the model. The model
16 overestimates PM_{2.5} concentrations observed at Manaus all year round, but particularly during
17 the dry season. This positive model bias may be due to several factors including a possible
18 overestimation of biogenic SOA over tropical forests and/or the model resolution, which is not
19 fully capturing the gradient in PM_{2.5} concentrations between the arc of deforestation and the
20 relatively undisturbed forest near Manaus.

21 In previous work we carried out a detailed model sensitivity analysis that accounted for the
22 uncertainty in the emissions (including biomass burning) and in the model processes such as
23 wet removal and dry deposition of aerosol (Lee et al., 2013). This analysis confirms that the
24 parametric uncertainty in modelled PM_{2.5} concentrations at these four stations is dominated by
25 the uncertainty in the biomass burning emissions flux in the dry season and by the yield of
26 biogenic SOA in the wet season, rather than the removal processes in the model.

27 Figure 4 demonstrates the differences in the spatial and temporal variability between the three
28 fire emission datasets, with different emissions capturing the observations better in different
29 months and locations. The model with GFED3 emissions captures the average seasonal
30 variability in PM_{2.5} observed at Alta Floresta (Fig. 4; $r^2=0.69$) and Porto Velho ($r^2=0.94$)
31 reasonably well. In particular, better simulating the peak in dry season concentrations at Porto
32 Velho than both FINN1 ($r^2=0.72$) and GFAS1 ($r^2=0.85$) emissions. However, PM_{2.5}
33 concentrations observed towards the end of the biomass burning season at Alta Floresta

1 (September – November) and Porto Velho (October – November) are not well captured by
2 GFED3 emissions, leading to larger biases at these sites (NMBF= -0.73 and -0.48, respectively)
3 than with FINN1 emissions (-0.51 and -0.41, respectively). At Santarem, the model with
4 GFED3 emissions underestimates observed PM_{2.5} concentrations throughout the dry season,
5 leading to a relatively large model bias and poor correlation with the observations (NMBF= -
6 0.76, $r^2=0.39$). Agreement with the observations at this site is improved with either FINN1
7 (NMBF= -0.11, $r^2= 0.76$) or GFAS1 (NMBF= -0.39, $r^2= 0.75$) emissions (discussed further in
8 Sect. 4.2).

9 If we consider the inter-annual variability in simulated and observed PM_{2.5} concentrations
10 (Figure S2), we find that the results are consistent with the evaluation of the simulated seasonal
11 cycle. The smallest bias between model and observations is with the FINN1 emissions (NMBF=
12 -0.22) compared to GFED3 (NMBF= -0.36) or GFAS1 (NMBF= -0.48). One notable point is
13 that the model with GFED3 emissions simulates the highest PM_{2.5} concentrations for the 2010
14 drought year, relative to the model with GFAS1 or FINN1 emissions, leading to improved
15 agreement with observations at Porto Velho (see Figs. 3a, 4a and S2).

16 In summary, the model captures the seasonal cycle and inter-annual variability of observed
17 PM_{2.5} reasonably well at biomass burning influenced sites in the Amazon. However, the model
18 underestimates observed concentrations in the dry season suggesting that the biomass burning
19 aerosol emission fluxes in all three emission inventories (GFED3, FINN1, GFAS1) may be
20 underestimated. We explore this further in Section 4.3.

21 **4.1.2 Aerosol optical depth in tropical biomass burning regions**

22 Figure 5 shows the simulated versus observed multi-annual monthly mean AOD at 440 nm at
23 each of the AERONET sites displayed in Fig. 1 (simulated and observed annual means are
24 compared in Fig. S3). Agreement between model and observed AOD is improved substantially
25 when biomass burning emissions are included in the model (Fig 5; NMBF= -0.40 to -0.18,
26 $r^2=0.62-0.69$) compared to the simulation without fire emissions (NMBF= -0.69, $r^2=0.22$). As
27 for PM_{2.5}, the bias in AOD across all sites is smallest with the FINN1 emissions (NMBF= -
28 0.18) compared to GFED3 (NMBF= -0.34) or GFAS1 (NMBF= -0.40). The model with FINN1
29 emissions also shows slightly improved correlation with the observations ($r^2=0.69$) relative to
30 GFED3 ($r^2=0.67$) and GFAS1 ($r^2=0.62$).

31 Figure 6a shows the NMBF and correlation between simulated and observed multi-annual
32 monthly mean AOD at the individual AERONET sites, grouped by region. In South America,

1 the bias in modelled AOD is smallest with the FINN1 emissions (mean NMBF= -0.47)
2 compared to GFED3 (-0.69) and GFAS1 (-0.89) emissions, which is consistent with
3 comparisons between modelled and observed PM_{2.5} in Amazonia (Sect. 4.1.1). In Indochina,
4 the model with FINN1 emissions also gives the smallest bias (mean NMBF= -0.02), relative to
5 GFED3 (-0.21) and GFAS1 (-0.23). In Africa, the model bias is smallest with GFED3 emissions
6 (mean NMBF= -0.78) compared to GFAS1 (-0.90) and FINN1 (-0.96). In Equatorial Asia, the
7 model bias is small and does not vary substantially between the different emission datasets
8 (FINN: 0.02, GFAS: -0.01, GFED: -0.02). In terms of temporal agreement between model and
9 observations, the correlation is noticeably stronger with GFED3 (mean $r^2=0.52$) in Africa and
10 with FINN1 (mean $r^2=0.75$) in Indochina, relative to the other emission datasets.

11 In general, the model with fire emissions captures the seasonal variability in observed AOD
12 best in South America (mean $r^2=0.90$) and captures the magnitude of observed AOD best in
13 Southeast Asia (Equatorial Asia: mean NMBF= -0.00; Indochina: mean NMBF= -0.14). The
14 agreement between model and observations in Africa is relatively poor, with substantial
15 underestimation of observed AOD (mean NMBF= -0.88). The negative model bias in Africa is
16 unlikely to be solely due to an underestimation of biomass burning aerosol and is likely
17 complicated by a contribution from dust (Pandithurai et al., 2001; Sayer et al., 2014; Cesnulyte
18 et al., 2014; Queface et al., 2011). There is better agreement between the model and observed
19 AOD at Ascension Island, which observes aged biomass burning aerosol from the African
20 continent (Sayer et al., 2014), with all three emission inventories (mean NMBF= -0.38,
21 $r^2=0.84$). This suggests that the model is able to capture outflow of biomass burning emissions
22 from Africa.

23 At the South American sites located in regions of high biomass burning activity associated with
24 deforestation fires (Abracos Hill, Rio Branco, Ji Parana SE and Alta Floresta), there is a small
25 improvement in the correlation with observed AOD with FINN1 ($r^2=0.96-0.98$) and GFAS1
26 ($r^2=0.94-0.97$) emissions relative to GFED3 ($r^2=0.79-0.88$). At these sites, AOD observed at
27 the tail end of the biomass burning season (~October-November) is better captured by GFAS1
28 and FINN1 than GFED3, leading to the improved correlation relative to GFED3. The model
29 with GFED3 is generally better able to capture observed AOD at the peak of the biomass
30 burning season (~August-September) than GFAS1 and FINN, which is largely due to relatively
31 high GFED3 emission estimates for the drought years 2007 and 2010 (see Fig. S1). These
32 results are consistent with comparisons with observed PM_{2.5} concentrations at Porto Velho and
33 Alta Floresta (Sect. 4.1.1).

1 At the AERONET sites located in Equatorial Asia and the Philippines (Singapore, Bandung,
2 Manila Observatory, ND Marbel Univ) an improved performance of either the GFAS1 or
3 GFED3 emission inventories may be expected over FINN1 (Andela et al., 2013) due to their
4 improved representation of tropical peatlands (in Indonesia and Malaysian Borneo) in their
5 biome maps (van der Werf et al., 2010). The agreement between AOD observed at Bandung,
6 Indonesia and the model is marginally improved with GFED3 (NMBF= -0.14, $r^2=0.52$) or
7 GFAS1 (NMBF= -0.15, $r^2=0.47$) relative to FINN1 (NMBF= -0.18, $r^2=0.34$). However, at the
8 other sites we find no strong indication of an improved performance with GFED3 (NMBF= -
9 0.06 to 0.13, $r^2=0.15-0.24$) or GFAS1 (NMBF= -0.03 to 0.14, $r^2=0.13-0.56$) relative to FINN1
10 (NMBF= 0.04 to 0.17, $r^2=0.16-0.42$). At most of these sites the model does not simulate a strong
11 contribution of biomass burning to AOD, likely due to their urban locations, which may explain
12 why we do not see a substantial difference in the performances of the three emission datasets.
13 Long-term ground-based retrievals of AOD located outside the influence of urban environments
14 are lacking in Equatorial Asia.

15 At the African AERONET sites, observed AODs are generally better captured by the model
16 with GFED3 emissions (mean NMBF= -0.78, $r^2=0.52$) than with FINN1 (mean NMBF= -0.96,
17 $r^2=0.35$) or GFAS1 (mean NMBF= -0.90, $r^2=0.41$) emissions. Andela et al. (2013) report that
18 the GFED3 emissions flux of carbon monoxide (CO) is higher than GFAS1 or FINN1 for humid
19 savannah regions, where the burned area product may observe more cloud covered fires than
20 active-fire detection. This feature may explain the improved simulation of AOD with GFED3
21 over Africa. Andela et al. (2013) also report that the FINN1 emission estimates of CO are lower
22 than both GFED3 and GFAS1 in global savannah regions, with the largest spatial deviation
23 found in humid savannahs where fire size is large. This may suggest that the assumed fire size
24 in FINN1 for savannah fires (0.75 km^2) could be too small for humid savannah fires in Africa,
25 contributing to an underestimation of AOD in this region.

26 **4.1.3 Overview of PM_{2.5} and AOD evaluation**

27 In the previous sections we have evaluated the model against ground based observations of
28 PM_{2.5} and AOD. In general, we find that the model is negatively biased against observations
29 in regions strongly influenced by biomass burning. However, the model bias in surface PM_{2.5}
30 concentrations is generally smaller than for AOD over South America, where observations of
31 both quantities are available ($\text{NMBF}_{\text{PM}_{2.5}} = -1.85$ to -0.25 , $\text{NMBF}_{\text{AOD}} = -2.38$ to -0.40 ; see Figs.
32 2 and S4). If we compare average model biases (with fires) in multi-annual monthly mean

1 PM2.5 and AOD (for 2003-2004) at locations where AERONET stations are in close proximity
2 to the PM2.5 measurement stations, we find a larger model bias in AOD at Santarem/Belterra
3 ($NMBF_{PM2.5} = -0.61$, $NMBF_{AOD} = -1.15$), but the reverse at Alta Floresta ($NMBF_{PM2.5} = -0.64$,
4 $NMBF_{AOD} = -0.42$).

5 These results suggest that although the negative model bias in PM2.5 and AOD may be partly
6 due to an underestimation of biomass burning aerosol emissions (due to uncertainties associated
7 with fire detection and subsequent calculations of emission fluxes), there are likely to be other
8 factors contributing to the model discrepancy in AOD that do not affect modelled surface
9 PM2.5 concentrations. These factors include uncertainties in the calculation of AOD that are
10 largely associated with assumptions made about the aerosol optical properties (assumed
11 refractive indices), mixing state (external/internal mixing) and hygroscopic growth of the
12 aerosol. We investigate the sensitivity of simulated AOD to these assumptions below.

13 As described in Sect. 3.2, to calculate AOD at 440 nm we use component-specific refractive
14 indices from Bellouin et al. (2011) for a wavelength of 468 nm ($1.500 - 0.000i$ for POM and
15 $1.750 - 0.452i$ for BC). To test the sensitivity of AOD to the choice of refractive indices, we
16 applied the refractive indices tested by Matichuk et al. (2007) for smoke aerosol ($1.54 - 0.025i$
17 calculated by Haywood et al. (2003) for young smoke aerosol over southern Africa; $1.51 -$
18 $0.024i$ and $1.52 - 0.019i$ retrieved by an AERONET station, Ndola in Zambia, located close to
19 smoke sources) to the BC and POM components in our model. We find that the modelled AOD
20 is relatively insensitive to the choice of complex refractive index within the range of values
21 tested here (altering the magnitude of AOD by less than 5%), which is in agreement with
22 Matichuk et al. (2007). Although the range of refractive indices tested is relatively narrow
23 (Matichuk et al., 2007), this result suggests that uncertainty in the assumed refractive indices is
24 unlikely to explain the discrepancy in simulated AOD.

25 We also find that the AOD is fairly insensitive to the mixing state assumption, with limited
26 difference in simulated AOD between assuming optical properties derived from an external
27 mixture of aerosol species and an internal (volumetrically-averaged) mixture. **Figure S5** shows
28 the simulated versus observed multi-annual monthly mean AOD at AERONET sites when
29 assuming external and internal mixing and indicates that the difference is less than 5%, with
30 internal mixing causing slightly higher AOD at the AERONET sites. However, we note that
31 the internal mixing assumption used in this study does not take into account the lensing effects
32 of coating BC with organic aerosol, which has been shown to interact with the aerosol
33 absorption in a non-linear way (Saleh et al., 2015).

1 As described in Sect. 3.2, the hygroscopic growth of the aerosol is calculated in the model using
2 the ZSR scheme. To test the sensitivity of AOD to aerosol hygroscopic growth, we instead use
3 the κ -Köhler water uptake scheme, based upon the Köhler equation with a single hygroscopic
4 parameter, κ , defining the water uptake for different chemical species (Petters and Kreidenweis,
5 2007) (see description of method in Sect. S1 of the supplementary material). For the SO_4 and
6 sea spray components in the model we used the mean values of κ for ammonium sulphate and
7 sodium chloride for subsaturated air masses (0.53 and 1.12, respectively) from Petters and
8 Kreidenweis (2007). BC is considered entirely hydrophobic in this model when using this
9 scheme. A wide range of κ values have been reported for organic aerosol (~0.01-0.25; Petters
10 and Kreidenweis, 2007) and biomass burning particles specifically (0.02-0.8; DeMott et al.,
11 2009; Petters et al., 2009). Engelhart et al. (2012) reported κ values of between 0.06 and 0.6 for
12 primary biomass burning aerosol in a smog chamber (fuels representative of North American
13 wildfires), with photochemical ageing reducing the range of κ values to 0.08 to 0.3, with
14 biomass burning SOA having κ values of 0.11. We assume a κ value for POM (0.1) based upon
15 aerosol samples, largely composed of SOA, collected at the Manaus ground station (TT34)
16 during the 2008 Amazonian Aerosol Characterization Experiment (AMAZE-08) (Gunthe et al.,
17 2009). We test the sensitivity of simulated AOD to different κ values for both SO_4 and POM.
18 Figure 7 shows a comparison between AOD simulated using ZSR and the κ -Köhler scheme.
19 Using the κ -Köhler scheme and κ defined above, the water uptake is reduced relative to the
20 ZSR scheme, reducing the simulated AOD on average by a factor of 1.6 (range 1.1 to 2.3) at
21 AERONET sites (see Figs. 7a and 7b). This large reduction relative to ZSR is in part from the
22 assumption that the SO_4^{2-} component behaves as ammonium sulphate rather than the more
23 hygroscopic sulphuric acid, and the reduced water uptake for POM. To explore the sensitivity
24 to assumed κ values we increased κ values separately for SO_4 and POM. Assuming a higher κ
25 for sulfate (1.19 as for sulphuric acid, Fig. 7c) results in simulated AOD being a factor 1.25
26 lower than ZSR. Assuming a higher κ for both sulfate (1.19) and for POM (0.2) results in
27 simulated AOD being a factor of 1.18 lower. Our results highlight the large uncertainty present
28 in the simulated AOD due to aerosol hygroscopicity. AOD simulated with ZSR (assuming
29 sulfuric acid and high water uptake for organics) appears to be an upper estimate for water
30 uptake. This result is confirmed by comparing simulated AOD and mass extinction efficiencies
31 for the two water uptake cases against observations and values from other global aerosol models
32 (see Sect. S2 and Table S2).

1 Calculated AOD is also sensitive to errors in relative humidity (Myhre et al., 2009), which are
2 here taken from ECMWF re-analysis. Since water uptake is not a linear function of RH,
3 calculated AOD will also be sensitive to spatial resolution of the aerosol and RH fields. Coarse
4 spatial resolution (here 2.8°) will not capture fine scale variability in RH that will influence
5 measurements from AERONET stations. A higher resolution model would be required to test
6 how sensitive the simulated AOD is to the spatial resolution of the aerosol and RH fields and
7 whether or not increasing the resolution improves the agreement with observed AOD (and
8 reduces the discrepancy between the model performance in AOD and PM_{2.5}). Bian et al. (2009)
9 showed that increasing the resolution of the RH field from $2^\circ \times 2.5^\circ$ to $1^\circ \times 1.25^\circ$ can increase
10 simulated AOD by ~10% in biomass burning regions. This suggests the coarse resolution of
11 our global models may partly explain the underestimation of AOD and the larger discrepancies
12 with observed AOD compared to PM_{2.5}.

13 Errors may also exist in the model representation of biomass burning aerosol, for example in
14 the modelled particle size distribution, altering simulated optical properties of the aerosol and
15 thus calculated AOD. In addition, since AOD is a column-integrated quantity, an
16 underestimation of AOD may be due to an underestimation of aerosol concentrations aloft since
17 we have shown that the model agrees relatively well with PM_{2.5} concentrations observed at the
18 surface.

19 Further uncertainties in the model representation of biomass burning aerosol are associated with
20 the conversion of OC to organic matter (OM), which would affect both PM_{2.5} concentrations
21 and AOD predicted by the model. Increasing the assumed OM:OC ratio would increase the
22 total simulated mass of biomass burning aerosol. In our model we assume a relatively low
23 OM:OC ratio of 1.4 compared to previous studies on biomass burning aerosol. Kaiser et al.
24 (2012) use a value of 1.5, but note this ratio is low compared to values of around 2.2 proposed
25 for aged pollution and biomass burning aerosols by Turpin and Lim (2001), Pang et al. (2006)
26 and Chen and Yu (2007) and a value of 2.6 used by Myhre et al. (2003) for biomass burning
27 aerosol in southern Africa. These larger OM:OC ratios could account for in-plume (sub-grid)
28 atmospheric oxidation and subsequent SOA formation observed in some biomass burning
29 plumes (Vakkari et al., 2014). In future work we need to include the formation of semi-volatile
30 SOA in biomass burning plumes that has been shown to be important (Konovalov et al., 2015;
31 Shrivastava et al., 2015).

1 **4.2 Small-scale fires**

2 The GFED3 fire emissions are known to underestimate contributions from small-scale fires
3 (smaller than ~100 ha) that are below the detection limit of the global burned area product
4 derived from MODIS (Randerson et al., 2012). However, many of these small fires generate
5 thermal anomalies that can be detected by satellites (Randerson et al., 2012). This means that
6 fire inventories using active fire detections to derive emissions (FINN1 and GFAS1) will better
7 capture these small fires (Kaiser et al., 2012). Kaiser et al. (2012) demonstrate that GFAS1
8 includes emissions from small fires that are omitted in GFED3. Some of the differences
9 between the spatial patterns of emissions seen in Fig. 1 are likely due to missing small fires in
10 GFED3.

11 This result is corroborated by our comparisons between modelled and observed PM_{2.5}
12 concentrations at Santarem in the north region of Brazil (Sect. 4.1.1), where the poor agreement
13 between the observations and model with GFED3 emissions (NMBF= -0.76, $r^2=0.39$) is
14 substantially improved by using either of the active-fire based emission inventories (FINN:
15 NMBF= -0.11, $r^2= 0.76$; or GFAS: NMBF= -0.39, $r^2= 0.75$). Randerson et al. (2012) show that
16 in the region surrounding the Santarem station there is a particularly high small fire fraction of
17 total burned area, which explains why the GFED3 emissions do not capture the observations in
18 this region of Brazil. This result is consistent with comparisons between modelled and observed
19 AOD at the nearby AERONET station, Belterra. At this station, the model better captures the
20 observed AOD with either FINN1 (NMBF= -0.85, $r^2=0.84$) or GFAS1 (NMBF= -1.02, $r^2=0.81$)
21 emissions than with GFED3 emissions (NMBF= -1.58, $r^2=0.29$).

22 The improved representation of small fire emissions in FINN1 and GFAS1 may also explain
23 the improved agreement between modelled and observed PM_{2.5} (Sect. 4.1.1) and AOD (Sect.
24 4.1.2) towards the end of the burning season (~October-November) in Amazonia. Kaiser et al.
25 (2012) report that GFAS1 exhibits slightly longer fire seasons in South America than GFED3.
26 Fires occurring at the tail end of the biomass burning season may be smaller in size and thus
27 better captured by using an active-fire based emission inventory (GFAS1 and FINN1
28 emissions). While at the peak of the burning season in Amazonia, when fires are potentially
29 larger, the comparisons in Sects. 4.1.1 and 4.1.2 suggest that GFED3 emissions capture the
30 observations better than FINN1 or GFAS1.

31 In Indochina, there is improved agreement between simulated and observed AOD with FINN1
32 emissions (Fig. 6a; NMBF= -0.26 to 0.19, $r^2=0.14-0.98$) relative to both GFED3 (NMBF= -

1 0.54 to -0.08, $r^2=0.11-0.84$) and GFAS1 (NMBF= -0.51 to -0.08, $r^2=0.03-0.83$). Figure 8
2 compares the model with different emissions against observations at the nine AERONET sites
3 in Indochina. FINN1 emissions lead to an improved correlation with observations at all sites
4 and a reduced root mean square model error at six sites compared to GFED3 and GFAS1. Figure
5 9 compares the multi-annual average seasonal cycle in AOD at four sites in Thailand. The
6 model with GFED3 and GFAS1 emissions underestimates AOD observed during the dry season
7 (~January – May), whereas the model with FINN1 emissions captures the magnitude of dry
8 season AOD reasonably well.

9 AERONET sites in Indochina (located in north and central Thailand and Vietnam) are
10 influenced by local agricultural burning (Li et al., 2013; Lin et al., 2013; Sayer et al., 2014) of
11 sugarcane and rice crop residues (Gadde et al., 2009; Sornpoon et al., 2014). Agricultural fires
12 are typically smaller than other fire types (e.g., deforestation, grassland/savannah and forest),
13 with burned areas of ~0.3 to ~16 ha reported for individual agricultural fires in the US (McCarty
14 et al., 2009) and Africa (Eva and Lambin, 1998). The prevalence of small fires in Indochina
15 may explain why FINN1 emissions result in better prediction of AOD compared to GFED3 in
16 this region.

17 We do not find an improved prediction of AOD with GFAS1 compared to GFED3 in this
18 region, although this would be expected since GFAS1 better captures emissions from small
19 fires than GFED3 (Kaiser et al., 2012). However, the GFAS1 FRP is converted to dry matter
20 burned using GFED3 data (Heil et al., 2010; Kaiser et al., 2012), which may lead to an
21 underestimation of small fire emissions in some regions. Conversely, FINN1 assumes a
22 relatively large burned area of 1 km² (100 ha) for individual agricultural fires and therefore may
23 overestimate emission fluxes in agricultural fire regions. However, since many small fires may
24 be undetected as fire hotspots by MODIS (due to factors such as the small size of the fires,
25 orbital gaps, persistent cloud cover and the timing of satellite overpass i.e. the potential to miss
26 fires events), by oversizing the area of individual burns, the FINN1 emissions may compensate
27 for missing fire detections in this region (B. Yokelson, personal communication, 2014).

28 **4.3 Scaling biomass burning emissions**

29 Previous model simulations, summarised in Table 2, underestimate AOD in regions impacted
30 by biomass burning. To improve simulation of AOD, these studies have scaled particulate
31 emissions from biomass burning (or aerosol concentrations) by a factor of 1.02 to 6. We have
32 found that our model with three different fire emission datasets also underestimates both PM_{2.5}

1 and AOD across tropical regions (although to a lesser extent in Southeast Asia). In this section
2 we explore the impact of scaling biomass burning emissions on simulated AOD and PM_{2.5}
3 concentrations. We performed two sensitivity simulations with each emission inventory where
4 we perturbed the biomass burning emission fluxes of BC and POM upwards by factors of 1.5
5 and 3.4 (as recommended for GFED3 and GFAS1 by Kaiser et al. (2012)).

6 Figures 3b and 3c show the NMBF and correlation between simulated and observed multi-
7 annual monthly mean PM_{2.5} concentrations for the two simulations with scaled biomass
8 burning emissions. The outcome of scaling the emissions by a factor of 1.5 depends on the site
9 location. At the sites strongly impacted by biomass burning, the model bias in PM_{2.5} is reduced
10 (FINN_{x1.5}: -0.16 to 0.08; GFED_{x1.5}: -0.67 to -0.15; GFAS_{x1.5}: -0.89 to -0.22) with little
11 change in the correlation. At the preserved forest site near Manaus, the positive model bias is
12 increased (FINN_{x1.5}: 1.33; GFAS_{x1.5}: 0.69; GFED_{x1.5}: 0.66). The outcome of scaling the
13 emissions by a factor of 3.4 depends on both the site location and the emission dataset. The
14 model bias is increased at all sites with FINN1 emissions (0.63-2.72), with mixed results for
15 GFED3 (-0.39 to 1.18) and GFAS1 (-0.16 to 1.25) emissions. Any scaling of the emissions
16 leads to an overestimation of PM_{2.5} at Manaus with all three emission datasets.

17 In summary, a scaling factor of 1.5 applied to the FINN1 emissions is adequate for the model
18 to capture surface PM_{2.5} concentrations observed in regions of high fire activity in the Amazon
19 region. In contrast, the GFAS1 emissions require a larger scaling factor (closer to 3.4) for the
20 model to capture surface PM_{2.5} observed at these sites.

21 The results of scaling the GFED3 emissions are more complex. Scaling GFED3 emissions by
22 a factor of 1.5, the model bias becomes relatively small at Alta Floresta (-0.36) and Porto Velho
23 (-0.15) but remains large and negative at Santarem (-0.67). Scaling the emissions by a factor of
24 3.4 reduces the model bias at Santarem (-0.39), but leads to an overestimation of PM_{2.5} at the
25 other three sites (0.33-1.18). At Santarem, scaling GFED3 emissions by a factor 3.4 only
26 marginally improves agreement with the observations; the correlation remains below 0.5 and
27 model bias remains negative (despite a positive model bias at the other sites). This is because
28 GFED3 emission fluxes in the peak biomass burning season months in the region of Santarem
29 (November and December) are very low or non-existent, likely due to an omission of small
30 fires (Sect. 4.2), thus there are very few emissions to scale. This result suggests that even by
31 scaling GFED3 emissions by a large factor it is still possible to underestimate PM from fires in
32 regions influenced by emissions from small fires.

1 Figures 6a and 6b show the NMBF and correlation between simulated and observed multi-
2 annual monthly mean AOD with scaled biomass burning emissions. For the model with GFAS1
3 emissions, scaling by a factor of 3.4 reduces the model bias at all but one site in Indochina,
4 Africa and South America (relative to the simulations without scaling or with a scaling factor
5 of 1.5), resulting in the best overall match to observed AOD in these regions. In Equatorial Asia
6 the scaling required to capture observed AOD depends on the site location (two sites require no
7 scaling and two sites require a scaling factor of either 1.5 or 3.4).

8 For GFED3 emissions, scaling by a factor of 3.4 results in the best overall match to observed
9 AOD in Africa and Indochina, but leads to an increased model bias at half the sites in South
10 America. However, even with a scaling factor of 3.4, the model with GFED3 emissions
11 continues to underestimate observed AOD in north Brazil (Belterra; NMBF= -0.94), indicating
12 that a large scaling factor does not fully compensate for the likely omission of small fire
13 emissions in this inventory (Sect. 4.2). The overall result of scaling GFED3 emissions in
14 Equatorial Asia is the same for GFAS1 emissions.

15 Scaling FINN1 emissions by a factor of 3.4 improves the agreement with observed AOD in
16 Africa (at all sites), but generally leads to overestimation and increased model bias at sites in
17 South America and Southeast Asia. Scaling FINN1 emissions by a factor of 1.5 is adequate to
18 capture observed AOD at the majority of sites in South America (mean NMBF= -0.16), with
19 no scaling required for the majority of sites in Indochina (mean NMBF= 0.02) and Equatorial
20 Asia (mean NMBF= 0.02).

21 We note that even with a scaling factor of 3.4 applied to the biomass burning emissions, the
22 model underestimates observed AOD at the African AERONET sites with all three fire
23 emission inventories (mean NMBF= -0.31). This may indicate that a larger scaling factor is
24 required to capture observations in this region. However, using a too high scaling factor is likely
25 to compensate for model error e.g. too efficient removal of aerosol or underestimation of dust
26 emissions, and therefore overestimate the contribution of biomass burning to AOD. The
27 potential for compensation errors with emission scaling is relevant for all three regions. For
28 example, in South America the model bias in AOD in the wet season (~December to May) is
29 increased at four or more sites when the FINN1, GFED3, and GFAS1 emissions are scaled by
30 a factor of 3.4, which may be an indication of compensation errors. Compensation errors are
31 also likely to be occurring when emissions are scaled by a factor of 3.4 at sites in urban locations
32 (see Table S1 for location classifications), where a global model is unable to capture sub-grid-
33 scale urban emissions.

1 5. Conclusions

2 Particulate emissions from open biomass burning (landscape fires) are very uncertain.
3 Numerous previous studies underestimate AOD in regions impacted by fires and scale
4 particulate emissions by up to a factor of 6 to match observed AOD (see Table 2). We have
5 used the GLOMAP global aerosol model evaluated against surface PM_{2.5} observations and
6 AERONET AOD to better understand particulate emissions from tropical biomass burning.

7 Simulated AOD is sensitive to a range of variables including i) vertical profile of aerosol, ii)
8 aerosol optical properties, chemical composition, size distribution and hygroscopic growth, iii)
9 relative humidity; and iv) model spatial resolution. In particular, we found that simulated AOD
10 is very sensitive to the calculation of hygroscopic growth, with the magnitude of AOD ranging
11 by a factor of ~1.6 between our upper and lower estimates. We assumed an upper estimate of
12 aerosol hygroscopic growth resulting in an upper estimate of AOD, reducing any emission
13 scaling required to match observed AOD.

14 We compared three different satellite-derived fire emission datasets (GFED3, GFAS1 and
15 FINN1). Total pan-tropical particulate emission (BC+OC) varied by less than 30% between the
16 different emission datasets. Regional differences were much larger (often exceeding 100%)
17 leading to important differences in aerosol concentrations simulated by the global model.

18 We found that GLOMAP underestimated both PM_{2.5} and AOD in regions strongly impacted
19 by biomass burning, with all emission datasets. The largest underestimation of AOD occurred
20 across Africa, which may be partly due to a large contribution of dust. The smallest
21 underestimation of AOD occurred over Equatorial Asia, where the contribution of fire
22 emissions to simulated AOD was also smallest. Overall, the smallest bias between model and
23 both PM_{2.5} and AOD observations was found using FINN1 emissions. The model with FINN1
24 emissions also best simulated the seasonal variability of AOD over Indochina, potentially
25 because of the dominance of smaller fires in this region that are better captured by the FINN1
26 dataset.

27 In South America where we have coincident surface PM_{2.5} and AOD observations,
28 underestimation of AOD is greater than underestimation of surface PM_{2.5}, even though we
29 assume upper estimates for aerosol water uptake. We suggest this discrepancy could be caused
30 by errors in the calculation of AOD (see above). We caution against using observations of AOD
31 to scale emissions before these issues are fully understood.

1 For each emission dataset we ran two additional simulations where we scaled emissions by
2 factors of 1.5 and 3.4, within the range of previous studies (Table 2). We find that the scaling
3 that results in the best agreement with observations is regionally variable and depends on the
4 emission dataset used. With FINN1 emissions, PM_{2.5} concentrations and AOD in South
5 America are well simulated when emissions are increased by 50%, whereas AOD in Africa is
6 more consistent with a factor 3.4 scaling. In Southeast Asia, observed AOD is well simulated
7 without any scaling applied; scaling FINN1 emissions by 50% generally leads to overestimation
8 in this region. With GFAS1 emissions, PM_{2.5} concentrations in South America and AOD in
9 South America, Africa and Indochina are best simulated when emissions are scaled by a factor
10 3.4. With GFED3 emissions, observations of PM_{2.5} in north Brazil and AOD in Africa,
11 Indochina and some regions of South America are also better simulated with a factor 3.4
12 scaling; for PM_{2.5} concentrations and AOD observed in active deforestation regions of South
13 America, a 50% scaling is sufficient. In Equatorial Asia, the results of scaling both GFAS1 and
14 GFED3 emissions are mixed and depend on site location; overall observed AOD is captured
15 best either without scaling or with a scaling factor of 1.5.

16 A factor 1.5 scaling is within the uncertainty of emission datasets and is substantially smaller
17 than the emission scaling applied by many other studies (see Table 2). We note that a factor 1.5
18 scaling is within the uncertainty of assumed OM to OC ratios; we assume an OM:OC ratio of
19 1.4 which is at the low end of other studies (Tsigaridis et al., 2014). We have treated biomass
20 burning emissions as primary and non-volatile. Formation of semi-volatile SOA in biomass
21 burning plumes may be important (Konovalov et al., 2015; Shrivastava et al., 2015) and needs
22 to be explored in future work. Scaling fire emissions by a factor of 3.4 to match AERONET
23 AOD is likely to partly compensate for an underestimation of aerosol from other sources e.g.
24 dust and/or urban emissions and may also compensate for errors in the calculation of AOD.
25 Simulated AOD is sensitive to a range of variables including relative humidity, spatial
26 resolution, aerosol hygroscopicity, aerosol size distribution, aerosol optical properties and
27 mixing state (external/internal mixing). In particular we found strong sensitivity of AOD to
28 aerosol hygroscopicity and water uptake, with simulated AOD at AERONET locations varying
29 by a factor of 1.6 between upper and lower estimates of aerosol hygroscopicity. We note that
30 there is the potential for compensating errors across these different uncertainties. Analysis
31 against detailed aerosol optical, microphysical and chemical measurements (Brito et al., 2014;
32 Andreae et al., 2015) is now required to further understand the issues raised here.

1 Problems with the detection of small fires are an acknowledged issue for GFED3, which relies
2 on detections of area burned to derive emissions (Randerson et al., 2012). Over regions that are
3 likely dominated by small fires the model with GFED3 emissions substantially
4 underestimates both PM_{2.5} (north Brazil) and AOD (north Brazil and Thailand). The model
5 with GFAS1 and FINN1 emissions better simulates aerosol in these regions providing
6 independent evidence that these datasets better represent emissions from small fires. We note
7 that the most recent version of GFED emissions (GFED4) includes an estimate of emissions
8 from small fires (Giglio et al., 2013). Future work should evaluate these emissions against
9 aerosol observations to assess the representation of small fire emissions in the specific regions
10 highlighted here.

11

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18

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1 **Table 1.** Summary of biomass burning emission inventories used in this study: the Global Fire
2 Emissions Database version 3 (GFED3), the National Centre for Atmospheric Research Fire Inventory
3 version 1.0 (FINN1) and the Global Fire Assimilation System version 1.0 (GFAS1). For each emission
4 inventory, the total amounts of black carbon (BC) and organic carbon (OC) aerosol emitted from fires
5 over the tropical region (defined as 23.5°N to 23.5°S) are given for the 2003 to 2011 average. Numbers
6 in parenthesis give the ratio to GFED3 emissions.

	GFED3	GFAS1	FINN1
Method	MODIS burned area & biogeochemical model	MODIS thermal anomaly product & fire radiative power	MODIS thermal anomaly product & assumed burned area
Spatial resolution	0.5° x 0.5°	0.5° x 0.5°	1 km x 1 km
Temporal resolution	Monthly (1997 – 2011) Daily (2003 – 2011)	Daily (2001 – 2015)	Daily (2002 – 2013)
Amount of OC emitted over tropics (Tg a⁻¹)	13.412	11.731 (0.87)	17.282 (1.29)
Amount of BC emitted over tropics (Tg a⁻¹)	1.705	1.532 (0.90)	1.724 (1.01)
OC:BC ratio over tropics	7.87	7.66	10.02
Reference	Van der Werf et al., 2010	Kaiser et al., 2012	Wiedinmyer et al., 2011

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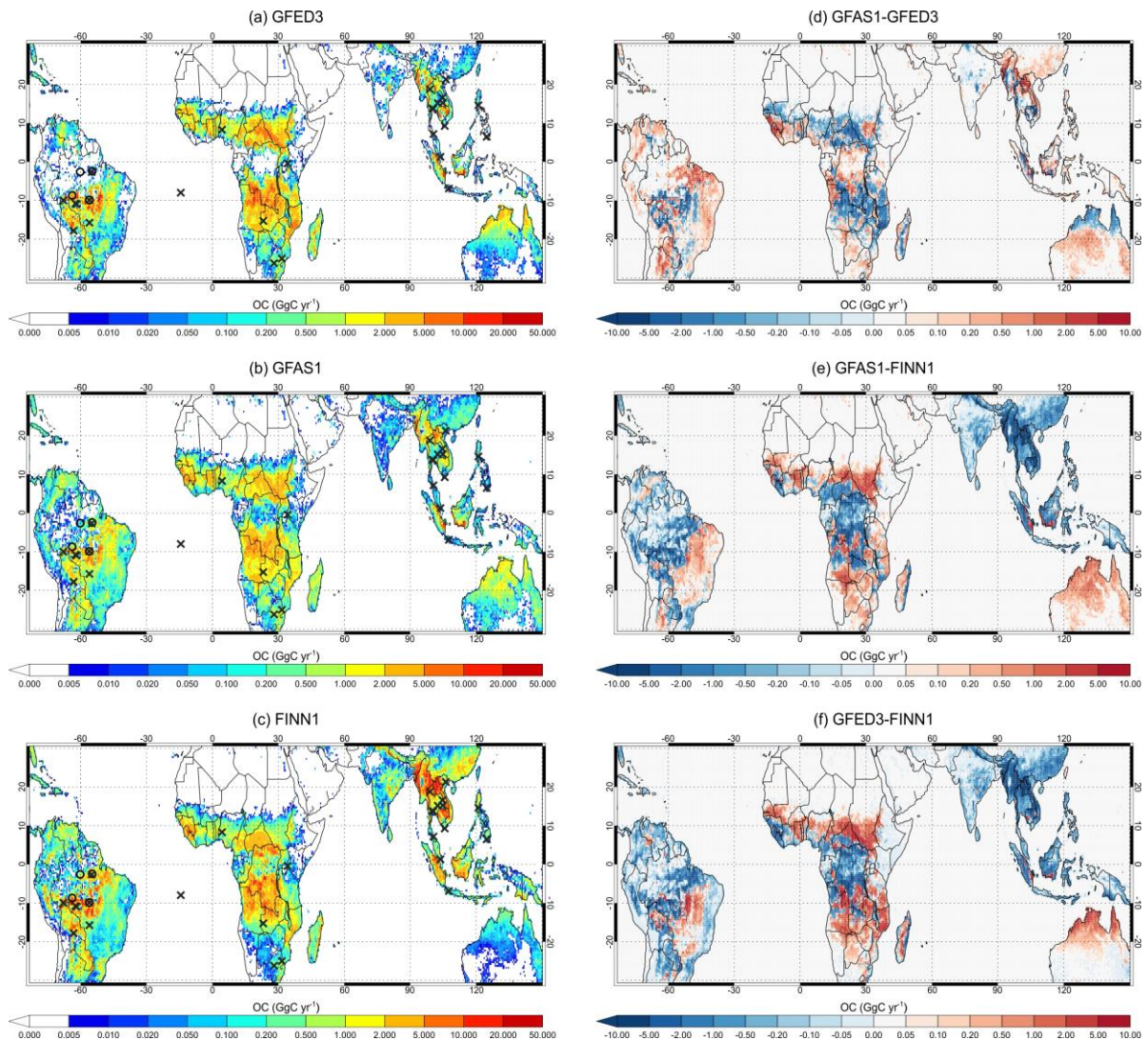
1 **Table 2.** Summary of scaling factors applied in previous modelling studies to biomass burning emissions
 2 or modelled concentrations of biomass burning aerosol to match observations. Region abbreviations
 3 used in the table are defined in van der Werf et al. (2006): Northern Hemisphere South America
 4 (NHSA), Southern Hemisphere South America (SHSA), Northern Hemisphere Africa (NHAF),
 5 Southern Hemisphere Africa (SHAF), Southeast Asia including the Philippines (SEAS) and Equatorial
 6 Asia (EQAS). See van der Werf et al. (2006; 2010) for discussion of differences between GFED versions
 7 1, 2 and 3; on average GFED3 are 13% lower than GFED2 van der Werf et al. (2010), with total GFED2
 8 emissions lower than GFED1 in Central and Southern America and Southern Africa (van der Werf et
 9 al., 2006).

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Reference	Biomass burning emission inventory	Region of focus	Details of scaling applied
Myhre et al., 2003	Biomass burning BC emissions from the Global Emissions Inventory Activity (GEIA), based on Cooke and Wilson (1996); OC emissions from Liousse et al. (1996).	Southern Africa	Used a relatively high OM/OC ratio of 2.6 and increased the modelled aerosol mass by 20% to account for mass fraction of inorganic components observed to be of 17% of the total mass.
Matichuk et al., 2007	GFED1 (van der Werf et al., 2003)	Southern Africa	Multiple sensitivity studies were performed with the model including simulations with halved and doubled fire emissions..
Matichuk et al., 2008	GFED2 (van der Werf et al., 2006)	South America	Smoke source function was scaled up by a factor of 6..
Johnson et al., 2008	Biomass burning emissions following Dentener et al. (2006): GFED1 (van der Werf et al., 2004) for year 2000 or a 5-year (1997–2001) average (not specified)	West Africa	Increased mass concentration of biomass burning AOD by a factor of 2.4..
Chin et al., 2009	Calculated using dry mass burned dataset from GFED2 (van der Werf et al., 2006)	Global	No scaling applied, but used emission factors of BC (1 g kg ⁻¹) and OC (8 g kg ⁻¹) that are 40–100% higher than commonly used values (Andreae and Merlet, 2001).
Sakaeda et al., 2011	Aerosol fields taken from MATCH chemical transport model	Southern Africa	OC and BC masses were increased by a factor of 2 over 10°N–30°S and 20°W–50°E.
Johnston et al., 2012	GFED2 (van der Werf et al., 2006)	Global	Scalar adjustments made for 14 continental scale regions: NHSA (2.48-2.7), SHSA (1.9-3.3), NHAF (1.02-1.08), SHSA (1.68-2.01), SEAS (2.43-3.08), EQAS (2.3-2.72). Scaling factors were applied to modelled surface fire PM2.5 to match satellite observations of AOD (non-fire aerosol was also scaled).
Kaiser et al., 2012	GFED3 and GFASv1.0	Global	Model was biased low in South America and Africa by factors of 4.1 and 3.0. Recommended a global

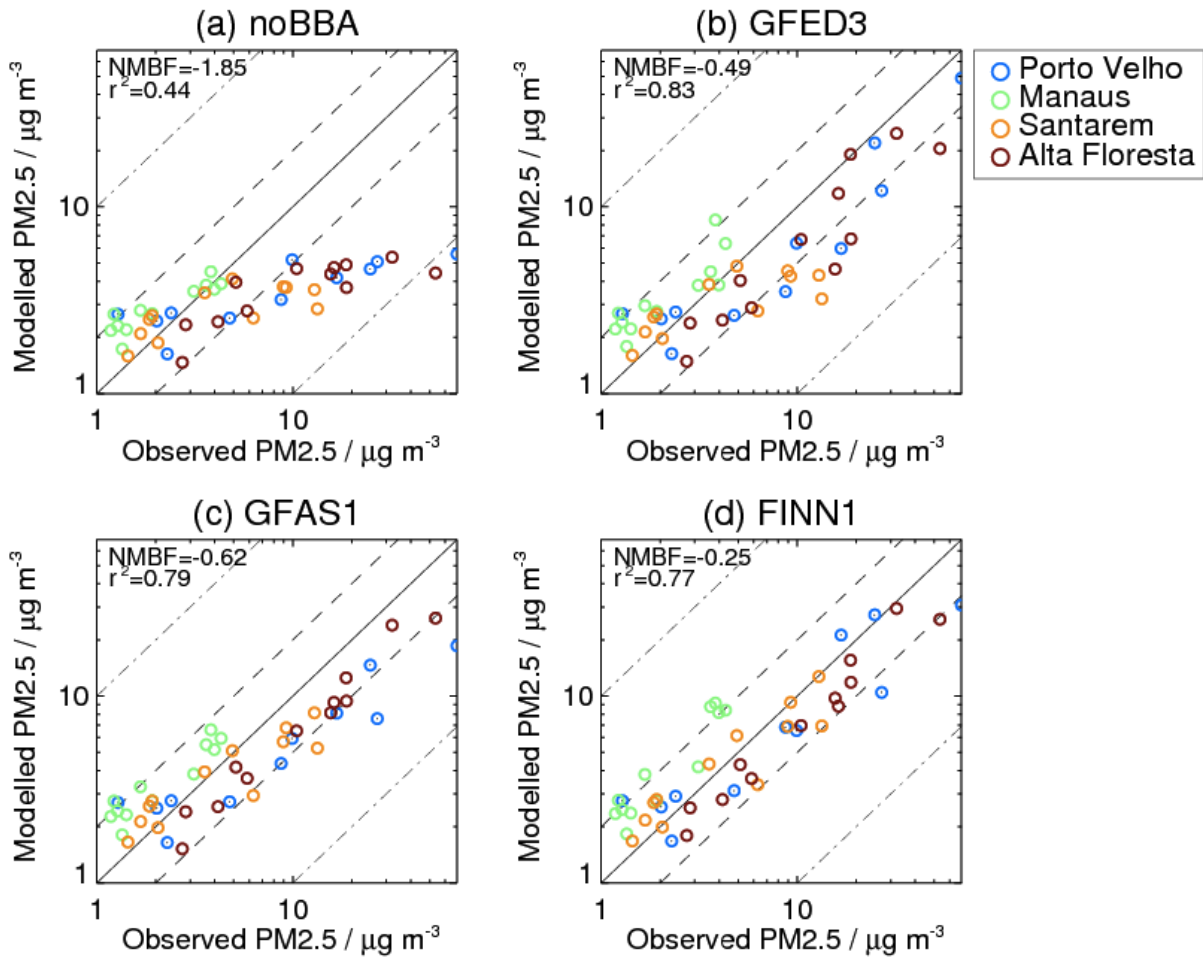
			enhancement of 3.4 for PM emissions from fires.
Ward et al., 2012	Calculated from Kloster et al. (2010, 2012) CLM3 simulations of global fire area burned; using emission factors from Andreae and Merlet (2001) and updates from Hoelzemann et al. (2004). Compared against GFED2.	Global	Scalar adjustments were made for continental scale regions following Johnston et al. (2012) with slight modifications: SHSA (2.0), NHAFF (1.0), SHAF (3.0), SEAS (1.5), EQAS (3.0). Scaling factor directly applied to model fire emissions.
Tosca et al., 2013	GFED3	Global	Biomass burning BC and OC emissions scaled by factor of 2 globally with additional regional scaling factors applied: South America (2.4), Africa (2.1), Southeast Asia (1.67).
Marlier et al., 2013	GFED3	Southeast Asia	Total aerosol burden scaled by 1.02-1.96 (depending on model), with additional scaling factors of 1.36-2.26 applied to fire aerosol..

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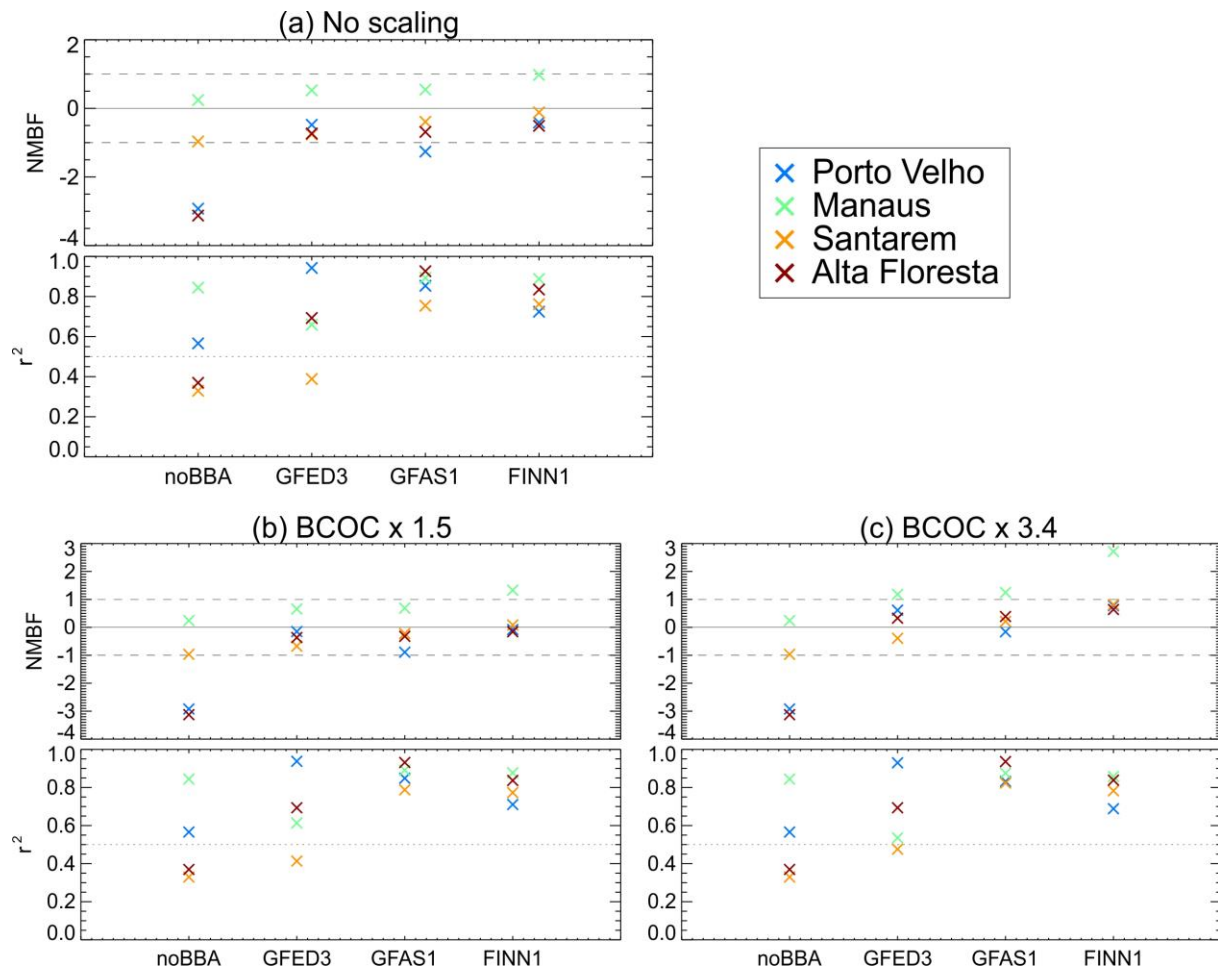
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2 **Figure 1.** (a)-(c) Total annual emissions of organic carbon (OC) in Gg(C) a^{-1} averaged over the period
 3 of January 2003 to December 2011 from (a) GFED3, (b) GFAS1 and (c) FINN1. Black circles mark the
 4 locations of the four aerosol measurement stations and black crosses mark the locations of the 27
 5 AERONET stations (see Table S1). (d)-(f) Absolute difference in 2003-2011 mean annual OC emissions
 6 between GFAS1, GFED3 and FINN1 (d) GFAS1 minus GFED3 (e) GFAS1 minus FINN1 (f) GFED3
 7 minus FINN1. The FINN1 OC emissions (with a $1 \text{ km} \times 1 \text{ km}$ horizontal resolution) were aggregated
 8 onto a grid of $0.5^\circ \times 0.5^\circ$ degree resolution to compare with GFED3 and GFAS1.

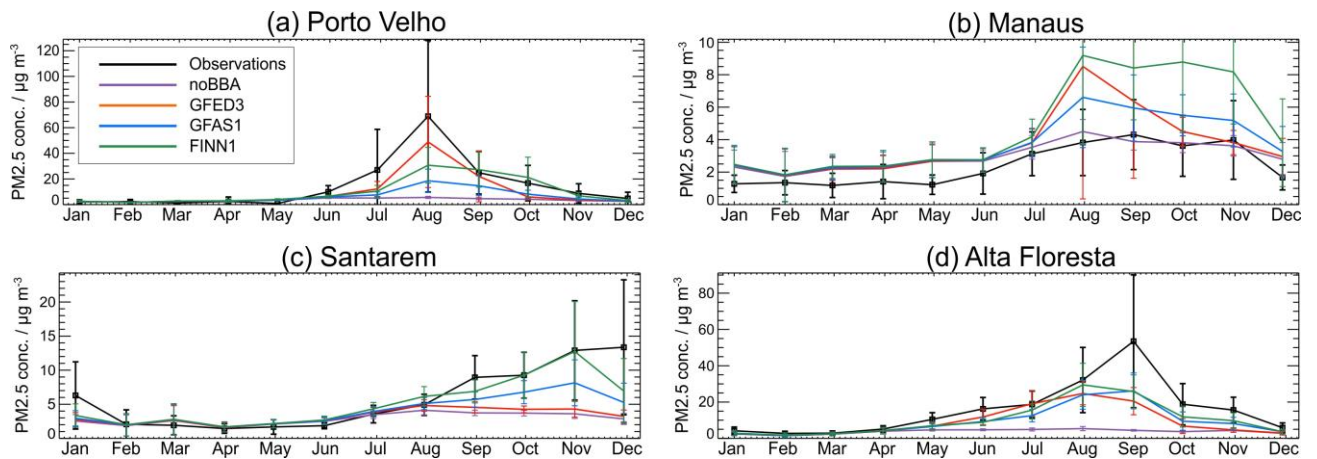


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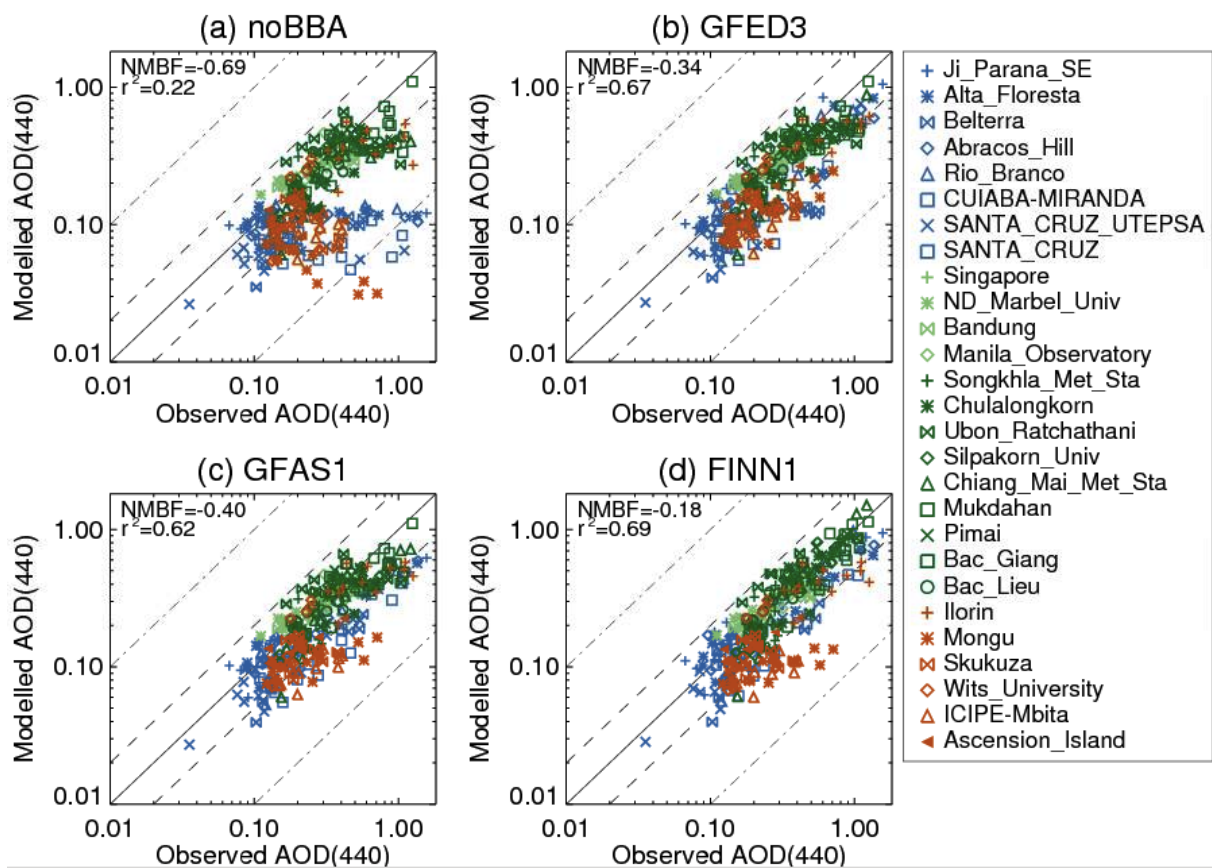
Figure 2. Simulated versus observed multi-annual monthly mean PM_{2.5} concentrations at each ground station in the Amazon region for the model (a) without biomass burning emissions, and with (b) GFED3, (c) GFAS1 and (d) FINN1 emissions. Multi-annual monthly mean concentrations were calculated by averaging over all years of data available between January 2003 and December 2011 to obtain an average seasonal cycle at each station. The normalised mean bias factor (NMBF; Yu et al., 2006) and Pearson's correlation (r^2) between modelled and observed PM_{2.5} concentrations are shown in the top left corner.



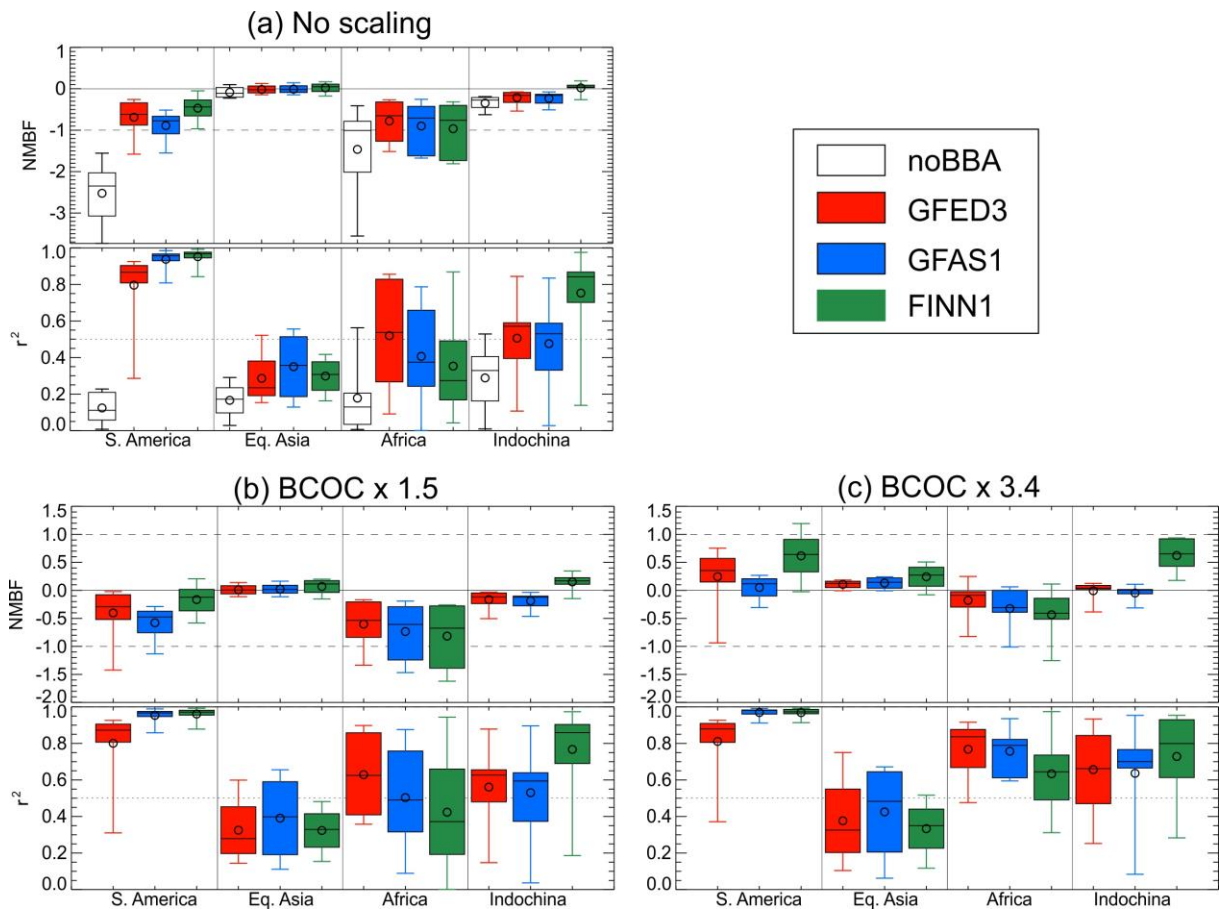
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2 **Figure 3.** Normalised mean bias factor (NMBF; Yu et al., 2006) and Pearson's correlation coefficient
3 (r^2) between modelled and observed multi-annual monthly-mean PM_{2.5} concentrations at each of the
4 four ground stations in Amazonia. Results are shown for four model simulations: without fires (noBBA),
5 and with each of the three biomass burning emissions inventories: GFED3, GFAS1, FINN1. (a) No
6 scaling applied to the fire emissions; (b) particulate (BC/OC) fire emissions scaled up globally by a
7 factor 1.5; (c) particulate (BC/OC) fire emissions scaled up globally by a factor of 3.4. The dashed lines
8 indicate NMBFs of -1 and 1, which equate to an underestimation and overestimation, respectively, of a
9 factor of 2. The dotted line indicates an r^2 value of 0.5.



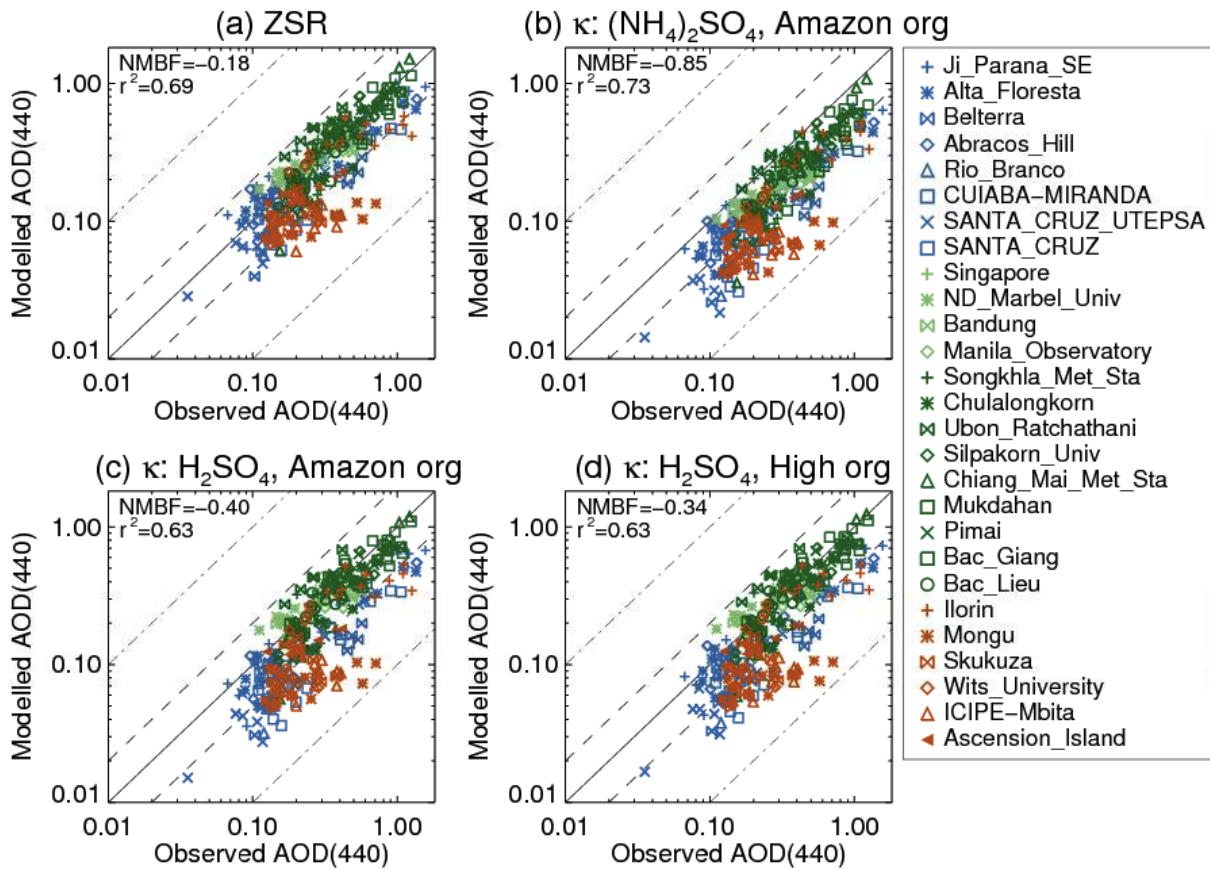
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 2 **Figure 4.** Average seasonal cycles in observed (black) and simulated (colour) multi-annual monthly
 3 mean PM_{2.5} concentrations at four ground stations in the Amazon region: **(a)** Porto Velho (2009-2011);
 4 **(b)** Manaus (2008-2011); **(c)** Santarem (2003-2006); and **(d)** Alta Floresta (2003-2004). Multi-annual
 5 monthly mean concentrations were calculated by averaging over all years of available observation data
 6 between January 2003 and December 2011. The modelled results are shown for four simulations:
 7 without biomass burning (purple), with GFED3 emissions (red), with GFAS1 emissions (blue) and with
 8 FINN1 emissions (green). The error bars show the standard deviation of the mean of the observed and
 9 simulated values, which represents the inter-annual and intra-monthly variability in the daily mean
 10 PM_{2.5} concentrations.



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 2 **Figure 5.** Simulated versus observed multi-annual monthly mean AOD at 440 nm at each AERONET
 3 station. The model is shown (a) without biomass burning emissions, and with (b) GFED3, (c) GFAS1
 4 and (d) FINN1 emissions. As for Fig. 2, the multi-annual monthly mean AODs were calculated using
 5 all years of daily mean data available between January 2003 and December 2011 to obtain an average
 6 seasonal cycle at each station. AERONET stations located in South America are shown in blue; stations
 7 in Southeast Asia are shown in green (stations in Equatorial Asia and Indochina in light and dark green,
 8 respectively); and stations in Africa are shown in orange. The normalised mean bias factor (NMBF) and
 9 Pearson's correlation (r^2) between modelled and observed PM_{2.5} concentrations are shown in the top
 10 left corner.

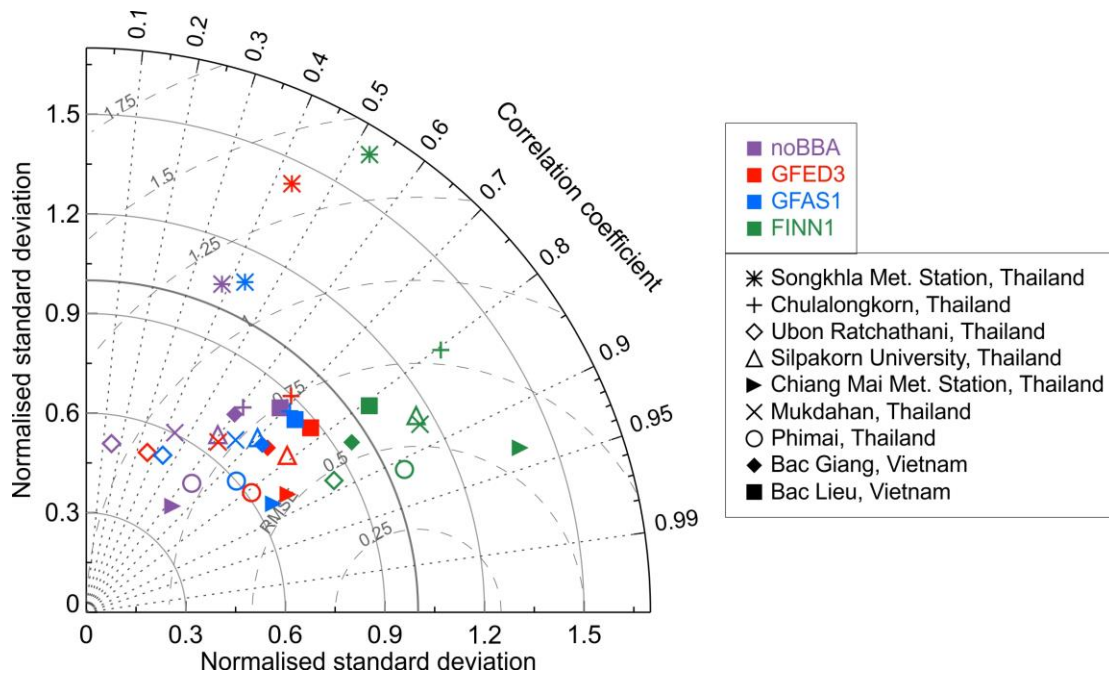


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 2 **Figure 6.** Box and whisker plots of the normalised mean bias factor (NMBF) and Pearson's correlation
 3 coefficient (r^2) between modelled and observed multi-annual monthly-mean AOD at 440 nm for
 4 AERONET stations located in South America (8 sites), Equatorial Asia (4 sites), Africa (6 sites) and
 5 Indochina (9 sites). Results are shown for four model simulations: without fires (white), and with each
 6 of the three biomass burning emissions inventories: GFED3 (red), GFAS1 (blue), FINN1 (green). (a)
 7 No scaling applied to the fire emissions; (b) particulate (BC/OC) fire emissions scaled up globally by a
 8 factor 1.5; (c) particulate (BC/OC) fire emissions scaled up globally by a factor of 3.4. The dashed lines
 9 indicate NMBFs of -1 and 1, which equate to an underestimation and overestimation, respectively, of a
 10 factor of 2. The dotted line indicates an r^2 value of 0.5.

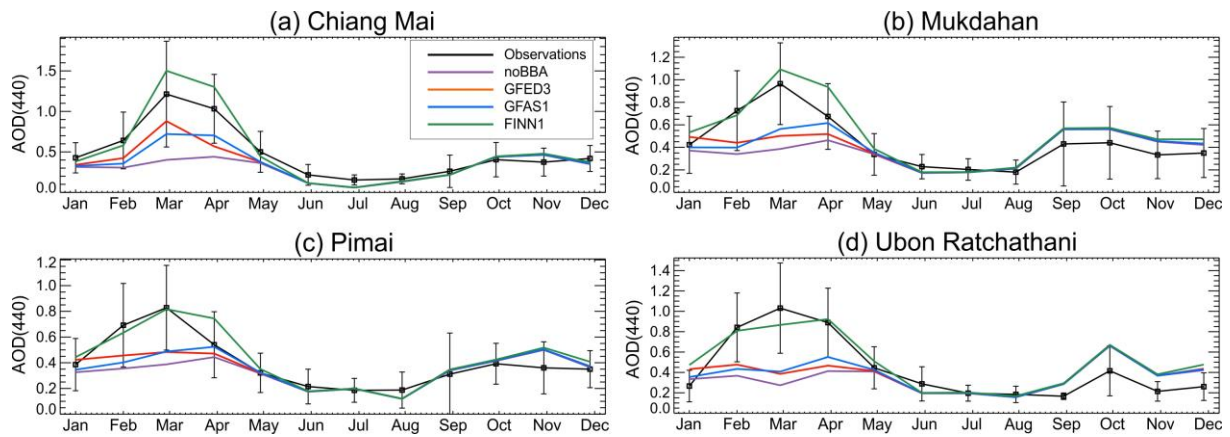


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2 **Figure 7.** Simulated versus observed multi-annual monthly mean AOD at 440 nm at each AERONET
3 station to demonstrate the sensitivity of simulated AOD to the calculation of aerosol water uptake. The
4 model is with FINN1 fire emissions and simulated AOD is calculated assuming internal mixing with (a)
5 ZSR water uptake scheme (identical to Fig. 5d); (b) κ -Köhler water uptake scheme: $\kappa_{\text{SO}_4}=0.53$, $\kappa_{\text{POM}}=0.1$;
6 (c) κ -Köhler water uptake scheme: $\kappa_{\text{SO}_4}=1.19$, $\kappa_{\text{POM}}=0.1$; and (d) κ -Köhler water uptake
7 scheme: $\kappa_{\text{SO}_4}=1.19$, $\kappa_{\text{POM}}=0.2$. AERONET stations located in South America are shown in blue;
8 stations in Southeast Asia are shown in green (stations in Equatorial Asia and Indochina in light and
9 dark green, respectively); and stations in Africa are shown in orange. The normalised mean bias factor
10 (NMBF) and Pearson's correlation (r^2) between modelled and observed PM_{2.5} concentrations are shown
11 in the top left corner.

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 2 **Figure 8.** Taylor diagrams (Taylor, 2001) comparing monthly mean modelled and observed AOD (440
 3 nm) at 9 AERONET stations located in Indochina. The modelled and observed monthly mean AODs
 4 were calculated for every month with available daily mean data between January 2003 and December
 5 2011. The observations are represented by a point on the x-axis at unit distance from the y-axis. The
 6 results are shown for four simulations: without biomass burning (purple), and with GFED3 (red),
 7 GFAS1 (blue) and FINN1 (green) fire emissions. The model standard deviation and root mean square
 8 error (RMSE) are normalised by dividing by the corresponding observed standard deviation. The
 9 normalised standard deviation and RMSE values are marked by the grey-solid and grey-dashed lines
 10 respectively. The correlation coefficient (r) values are marked by the grey dotted lines.



1
 2 **Figure 9.** Average seasonal cycles in observed (black) and simulated (colour) monthly mean AOD at
 3 440 nm at four AERONET stations in the Thailand: **(a)** Chiang Mai Met. Station; **(b)** Mukdahan; **(c)**
 4 Phimai; and **(d)** Ubon Ratchathani. Multi-annual monthly mean concentrations were calculated by
 5 averaging over all years of available daily mean observation data between January 2003 and December
 6 2011. The modelled results are shown for four simulations: without biomass burning (purple), and with
 7 GFED3 (red), GFAS1 (blue) and FINN1 (green) fire emissions. The error bars show the standard
 8 deviation of the mean of the observations.