1 Diagnosing uncertainties in global biomass burning emission inventories and

2 their impact on modeled air pollutants

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14 Abstract

15 Large uncertainties persist within current Biomass burning (BB) inventories, and the choice of 16 these inventories can substantially impact model results when assessing the influence of BB aerosols 17 on weather and climate. We evaluated discrepancies among BB emission inventories by comparing carbon monoxide (CO) and organic carbon (OC) emissions from seven major BB regions globally 18 19 between 2013 and 2016. Mainstream bottom-up inventories, including Fire INventory from NCAR 20 1.5 (FINN1.5) and Global Fire Emissions Database version 4s (GFED4s), along with top-down 21 inventories Quick Fire Emissions Dataset 2.5 (QFED2.5) and VIIRS-based Fire Emission Inventory 22 version 0 (VFEI0), were selected for this study.

23 Global CO emissions range from 252 to 336 Tg, with regional disparities reaching up to a sixfold 24 difference. Dry matter is the primary contributor to the regional variation in CO emissions (50-80%), 25 with emission factors accounting for the remaining 20-50%. Uncertainties in dry matter often arise 26 from biases in calculating bottom fuel consumption and burned area, influenced by vegetation classification methods and fire detection products. In the tropics, peatlands contribute more fuel 27 28 loads and higher emission factors than grasslands. At high latitudes, increased cloud fraction 29 amplifies the discrepancy in estimated burned area (or fire radiative power) by 20%. The global OC 30 emissions range from 14.9 to 42.9 Tg, exhibiting higher variability than CO emissions due to the 31 corrected emission factors in OFED2.5, with regional disparities reaching a factor of 8.7.

32 Additionally, we applied these BB emission inventories to the Community Atmosphere Model version 6 (CAM6) and assessed the model performance against observations. Our results suggest 33 34 that the simulations based on the GFED4s agree best with the MOPITT-retrieved CO. While 35 comparing the simulation with Moderate Resolution Imaging Spectroradiometer (MODIS) and AErosol RObotic NETwork (AERONET) aerosol optical depth (AOD), our results reveal that there 36 37 is no global optimal choice for BB inventories. In the high latitudes of the Northern Hemisphere, 38 using GFED4s and QFED2.5 can better capture the AOD magnitude and diurnal variation. In equatorial Asia, GFED4s outperform others in representing day-to-day changes, particularly during 39 40 intense burning. In Southeast Asia, we recommend using the OC emission magnitude from FINN1.5 combined with daily variability from QFED2.5. In the Southern Hemisphere, the latest VFEI0 has 41 42 performed relatively well. This study has implications for reducing the uncertainties in emissions or improving BB emission inventories in further studies. 43

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47 1 Introduction

In recent years, extreme wildfire events have occurred frequently around the world (Balshi et al., 48 2009; Knorr et al., 2016; Yang et al., 2019; Junghenn Noyes et al., 2022). The size of the fire has 49 50 consistently broken records over the last decades (Westerling et al., 2006; Westerling and Bryant, 2008; Brando et al., 2020), threatened lives and infrastructure, and continuously jeopardized the 51 global economy. Wildfires are also one of the most important sources of biomass burning (BB) 52 53 emissions, which can emit loads of gaseous and particulate pollutants (Ferek et al., 1998; Adams et 54 al., 2019), detrimental to regional air quality and human health (Reid et al., 2005, Reid and Mooney, 55 2016). Additionally, BB aerosols, predominantly black carbon (BC) and organic carbon (OC) can 56 affect regional climate by absorbing/scattering solar radiation, acting as cloud condensation nuclei, 57 and altering cloud albedo (Spracklen et al., 2011; Boucher et al. 2013). Recent studies have shown 58 that aerosols produced by biomass burning can significantly affect changes in temperature, cloud 59 fraction, precipitation, and even the circulation structure (Christian et al., 2019; Yang et al., 2019; Yu et al., 2019; Carter et al., 2020; Jiang et al., 2020; Ding et al., 2021; Huang et al., 2023). However, 60 61 these changes in meteorology are sensitive to the choice of BB emission inventory.

62 Previous studies often found that there is a significant deviation between the gaseous or particulate pollutants simulated by the model and the satellite retrieval value (Bian et al., 2007; 63 Chen et al., 2009; Carter et al., 2020), one of the most important reasons comes from the 64 65 uncertainties in emission inventories. For example, Bian et al. (2007) applied six different BB emission inventories, GFED1 and GFED2 (Global Fire Emissions Database version 1 and 2) 66 (GFED1 and GFED2), Arellano1, Arellano2, Duncan1, and Duncan2, to the Unified Chemistry 67 68 Transport Model (UCTM). They reported that although the total global CO of the six BB emission inventories was within 30% of each other, the model results suggested that regional deviations can 69 70 be much higher, by as much as 2-5 times, especially in the Southern Hemisphere. Bias in emission 71 inventories can therefore often have a significant impact on the direct and indirect effects of models 72 on aerosol assessments (Liu et al., 2018; Liu et al., 2020a; Ramnarine et al., 2019; Carter et al., 73 2020). Carter et al. (2020) compared the simulated black carbon (BC) and organic carbon (OC) 74 concentrations with measurements from IMPROVE (Interagency Monitoring of Protected Visual 75 Environments) observation network from May to September. They suggested that using the 76 FINN1.5 inventory (Fire INventory from NCAR 1.5) improves model results in eastern North 77 America, while using GFED4s, QFED2.4 (Quick Fire Emissions Dataset 2.4), and GFAS1.2 (Global Fire Assimilation System 1.2) inventories shows better agreement with observations in western 78 79 North America. They also noted that population-weighted BB PM2.5 concentrations in Canada and the adjacent United States could vary between 0.5 and 1.6 μ g m⁻³ in 2012 by using different BB 80 81 emissions. Liu et al. (2018) used the global model CAM5 (The Community Atmosphere Model 5) and three different BB emission inventories to analyze the uncertainties in the aerosol radiative 82 83 effects in the Northeastern United States in early April 2009. They found that aerosols exhibited a stronger cooling effect when CAM5 used the QFED2.4 inventory than the GFED3.1 and GFED4s 84 inventories, with additional cooling of -0.7 W m⁻² and -1.2 W m⁻² through aerosol direct radiative 85 effect and the aerosol-cloud radiative effect, respectively. On a global basis, Ramnarine et al. (2019) 86 87 used the global model GEOS-Chem-TOMAS (GEOS-Chem-TwO-Moment Aerosol Sectional), and 88 found that the direct radiative effects and indirect effects of aerosols driven by the FINN1.5 emission 89 inventory in 2010 were 70% and 10% lower than those driven by GFED4, respectively. Therefore,

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90 to better estimate regional aerosol-radiation/aerosol-cloud interactions in wildfire regions, it is

91 necessary to understand the differences in emission inventories from biomass combustion and the92 main drivers of uncertainties.

93 In general, BB emission inventories are based on bottom-up or top-down methods to infer the 94 emission source intensity. The bottom-up approach, also known as the fire detection and/or burned 95 area method, estimates emissions based on surface data such as fuel loading, active fire counts, 96 and/or burned area. Currently, the widely used BB inventories based on the bottom-up approach 97 include Duncan (Duncan, 2003), GFED (van der Werf et al., 2006, 2010a, 2010b, 2017), FINN 98 (Wiedingmyer et al., 2011), Global Inventory for Chemistry-Climate Studies-GFED4S (G-G) 99 (Mieville et al., 2010). The top-down approach uses satellite observations of fire radiative power 100 (FPR), a method to measure the radiative energy release rate of burning vegetation, to estimate 101 emissions by fuel consumption. The BB inventories based on the top-down method include Arellano 102 (Arellano Jr et al., 2004; Arellano Jr and Hess, 2006), GFAS (Kaiser et al., 2012), Fire Energetics 103 and Emission Research (FEER) (Ichoku and Ellison, 2014), QFED (Darmenov et al., 2015), the 104 Fire Emissions Estimate Via Aerosol Optical Depth (FEEV-AOD) (Paton-Walsh et al., 2012) and 105 the recently released VIIRS-based Fire Emission Inventory version 0 (VFEI0) (Ferrada et al., 2022). On a global scale, the average annual BB emissions of CO and OC can differ by a factor of 3 to 4, 106 with the global emissions fluctuating in the range of 280-580 Tg yr⁻¹ and 13-50 Tg yr⁻¹ respectively. 107 The bias may be even greater when focusing on emissions in specific regions (Bian et al., 2007; 108 109 Liousse et al., 2010; Williams et al., 2012; Carter et al., 2020; Lin et al., 2020b; Liu et al., 2020b). 110 For example, the estimated CO emission of Arellano inventory in South America during the burning 111 peak season of September 2000 is four times greater than that of GFED1 inventory (Bian et al., 2007). A recent study even found that since 2008, OC emissions from QFED2.5 in the Middle East 112 113 are approximately 50 times larger than those from GFED3 and GFED4 (Pan et al., 2020).

114 Several previous studies have analyzed the reason for the huge emission bias. According to 115 Darmenov et al. (2015), the emissions E_i (mass of pollutant i) is the sum of the products of the emission factor (EF) and the dry matter (DM) for each biome. While earlier studies suggested that 116 117 the uncertainty in BB emissions arises mainly from differences in emission factors (e.g., Alvarado 118 et al., 2010; Akagi et al., 2011; Urbanski et al., 2011), more recent studies point out that uncertainty 119 in dry matter also plays an important role (Paton-Walsh et al., 2010; 2012; Carter et al., 2020). For 120 example, Paton-Walsh et al. (2012) assessed the difference in CO emissions from the February 2009 Australian fire and found that total CO emissions in GFED3.1 were roughly three times higher than 121 122 that in FINN1, with DM contributing up to 80%. Carter et al. (2020) evaluated emissions from 123 various North American BB inventories over the period 2004-2016 and found that changes in DM 124 were very close to the emission trend, suggesting that uncertainty in potential DM across North 125 America was the primary factor, rather than EF.

126 The accuracy of BB inventories is influenced by land cover and land use (LULC) data, impacting both EFs and DM (Wiedinmyer et al., 2006; Ferrada et al., 2022). In a study by Wiedinmyer et al. 127 128 (2006), three distinct LULC products were employed to drive a regional BB emissions model. The 129 variations in LULC products led to discrepancies in fuel consumption, resulting in an annual bias 130 of up to 26% in North and Central America. Moreover, EFs are closely tied to different biomes, introducing uncertainty into BB emission inventories with varied biome classifications (Ferrada et 131 al., 2022). In addition to LULC products, uncertainties are introduced by fire detection products 132 133 (such as FRP and burned area products), affected by factors such as satellite transit time and cloud obscuration. For example, Paton-Walsh et al. (2012) found that in an Australian fire called "Black
Friday" in February 2009, the burned areas of FINN1 were barely half of that of GFED3.1. Liu et
al. (2020b) reported that compared with the active fire area used in FINN1.5, the burned area product
selected by GFED4s is less sensitive to the satellite overpass time and cloud obscuration. These
results indicate that LULC and fire detection products are key factors leading to bias in BB emission
estimation.

140 Although previous work has generated biomass burning emission inventories and attempted to reduce their uncertainties (Duncan, 2003; Arellano Jr et al., 2004; Arellano Jr and Hess, 2006; van 141 142 der Werf et al., 2006, 2010a, 2010b, 2017; Bian et al., 2007; Mieville et al., 2010; Wiedingmyer et 143 al., 2011; Kaiser et al., 2012; Paton-Walsh et al., 2012; Ichoku and Ellison, 2014; Darmenov et al., 2015; Liu et al., 2018; Ramnarine et al., 2019; Carter et al., 2020; Lin et al., 2020b; Liu et al., 2020b; 144 145 Pan et al., 2020; Zhang et al., 2020; Ferrada et al., 2022), they did not analyze the reasons why DM and EF exhibited large differences among various emission inventories, which may vary over time 146 and location. Here, this study aims to explore the underlying reasons for the differences in BB 147 148 emission inventories in major combustion regions around the world, thereby attempting to reduce 149 the uncertainties of the impact of BB emission inventories on model results. To minimize the interference of anthropogenic emissions on model results, we selected combustion regions 150 151 satisfying the following conditions: (1) regional BB CO emissions above 20 Tg yr⁻¹; (2) BB CO emissions contribute more than 70% of the total. We ultimately selected seven major burning areas 152 153 as shown in Fig. 1, including Boreal North America (BONA), Southern Hemispheric South America 154 (SHSA), Northern Hemispheric Africa (NHAF), Southern Hemispheric Africa (SHAF), Boreal Asia 155 (BOAS), Southeast Asia and India (SEAS), and Equatorial Asia (EQAS).

156 In this study, we compare several widely used datasets (FINN1.5, GFED4s, and QFED2.5) and 157 the recently released VFEI0. The former two datasets are based on the bottom-up method, while the 158 latter two are based on the top-down method. Specific details of these BB inventories are described in Section 2. In section 3, we explore the differences in CO and OC emissions among the four 159 160 inventories, examining the contributions of DM and EFs to these differences, respectively. For the 161 first time, we evaluate the biases of CO column concentrations and AOD driven by BB inventories 162 in the CESM2-CAM6 model. Based on our findings, we provide recommendations on which 163 inventory should be adopted across various regions. Section 4 presents the conclusion and 164 discussion, and our research is expected to offer insights into reducing the uncertainties with BB 165 emission datasets.

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167 2 Data and Methodology

168 2.1 Biomass Burning emission inventories

We simultaneously diagnosed the differences between two bottom-up approach inventories and two top-down approach inventories, including FINN1.5, GFED4s, and QFED2.5, which are commonly used in the current atmospheric model, as well as the recently released VFEI0. Details about the emission inventories and the satellite products they use are listed in Table 1 and Text S1 in supplementary.

174 Bottom-up (Burned Area) inventories

In this study, both FINN1.5 and GFED4s adopt a bottom-up approach (also called the BurnedArea method), and the details are shown in Table 1. FINN1.5 uses the MODIS (Moderate Resolution

177 Imaging Spectroradiometer) product MCD14DL for burned area calculations. This active fire 178 detection product monitors real-time fire points larger than 0.05 km². However, it is important to 179 note that if a fire occurs when the satellite is not in transit or is obscured by clouds during transit, it 180 will not be detected (Firms, 2017). Additionally, FINN1.5 assumes that every fire detected at the 181 equator (30°N-30°S) will persist the next day at half the size of the previous day (Table 1). However, 182 this assumption may not accurately reflect real-world conditions (Wiedinmyer et al., 2011; Pan et 183 al., 2020). The land cover classification in FINN1.5 is based on MCD12Q1 (IGBP, version 2005). 184 According to the IGBP land cover classification, each fire is initially assigned to one of 16 land 185 use/land cover (LULC) classes, and then lumped into six generic categories including tropical forest, 186 temperate forest, boreal forest, savanna and grasslands, woody savannas and shrublands, and 187 cropland (Fig. S1, Wiedingmyer et al., 2011). Emission factors (EFs) for various gaseous and particulate species are determined from a dataset compiled by Akagi et al. (2011) and Andreae and 188 Merlet (2001), with these EFs varying for different LULC types. Currently, FINN1.5 provides daily 189 190 global emissions from biomass burning since 2002, including 41 species, with a spatial resolution 191 of 1 km² (Table 1).

192 GFED4s differs in that it primarily uses the MCD64A1 Collection 5.1 burned area product (Giglio 193 et al., 2013; Randerson et al., 2018), capable of detecting fires larger than 500 m × 500 m. For small 194 fire areas, GFED4s incorporate active fire detection products (MOD14A1 and MYD14A1), compensating to some extent for the lower spatial resolution of the original product MCD64A1 (van 195 196 der Werf et al., 2017). In general, burned area products reduce uncertainty in fire detection due to 197 satellite non-transit and cloud/smoke obscuration when a burn occurs by identifying day-to-day 198 surface variations, such as charcoal and ash deposition, vegetation migration, and changes in vegetation structure (Boschetti et al., 2019). Similar to FINN1.5, each fire in GFED4s is initially 199 200 assigned to one of 16 LULC subcategories and then lumped into six categories, with the inclusion 201 of an additional biome, peatland (Fig. S1). EFs for various species follow Akagi et al. (2011) and 202 Andreae and Merlet (2001), varying across different biome categories. Currently, GFED4s provide 203 daily global emissions from biomass burning since 1997, including 27 species, with a spatial 204 resolution of $0.25^{\circ} \times 0.25^{\circ}$ (Table 1). However, since 2017, the DM provided by GFED4s is derived 205 from a linear relationship between past emissions and MODIS FRP data for the period 2003-2016. 206

207 Top-down (Fire Radiative Power) inventories

208 In this study, both QFED2.5 and VFEI0 use a top-down approach known as the Fire Radiative 209 Power (FRP) method. In contrast to the bottom-up approach, the top-down approach relies on 210 satellite products detecting fire-radiated power rather than fire point detection. QFED2.5 uses 211 MODIS Collection 6 MOD14/MYD14 level 2 products to estimate fire radiative power and pinpoint 212 fire locations using MOD03/MYD03 (Darmenov and Silva 2015; Liu et al., 2020b). The FRPs are 213 integrated over time to obtain fire radiative energy (FRE), which is converted to DM using an 214 empirical coefficient α. The initial α values are obtained from Kaiser et al. (2009) and are adjusted 215 monthly based on global emissions of GFED2 in 2003-2007. QFED2.5 classifies land cover using the International Geosphere-Biosphere Programme (IGBP-INPE) dataset, aggregating 17 land 216 217 cover classes into four broad vegetation types (Fig. S1, Darmenov and da Silva 2015). Initially, EFs 218 for various species in QFED2.5 also follow Akagi et al. (2011) and Andreae and Merlet (2001). But 219 for certain species, including organic carbon (OC), black carbon (BC), ammonia (NH₃), sulfur

dioxide (SO₂), and particulate matter diameter < 2.5μ m (PM_{2.5}), QFED2.5 incorporates a scaling factor to enhance the EFs. QFED2.5 provides daily global BB emissions since 2000, including 17 species, with a spatial resolution of $0.1^{\circ} \times 0.1^{\circ}$ (Table 1).

223 VFEI0 also adopts the top-down method but uses VNP14IMG.001 FRP product from VIIRS I-224 band (Visible Infrared Imaging Radiometer). This product has a higher resolution (375 m at nadir) 225 compared to MODIS (1 km resolution at nadir), enabling the detection of smaller and colder flames 226 (Ferrada et al., 2022). VFEI0 uses an empirical coefficient α derived from the linear regression of 227 GFED3.1 DM and VIIRS FRP to convert detected FRE into DM. VFEI0 uses MCD12C1 (IGBP, 228 version 2015) as the underlying LULC data, supplemented by Köppen climate classification (Beck et al., 2018), defining ten subcategories in VFEI0 (Fig. S1). VFEI0 groups these subcategories into 229 230 six biomes, corresponding to EFs provided by Andreae (2019). Currently, VFEI0 offers daily BB 231 emission since 20 January 2012, covering 46 emitted species with a horizontal resolution of 0.005° × 0.005° (Table 1). 232

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234 **2.2** The calculation for EFs and DMs

To calculate regional EFs and DMs, we adopt the approach outlined by Carter (2020). Initially,
we divide CO emissions per grid by the EF applied to each biome, yielding DM:

 $DM_{b,x} = CO_{b,x} / EF_b \tag{1}$

where b represents one of the seven biomes in Fig. S1, and x represents the location grid. This calculation of DM using CO is reasonably representative, given that the inventories are not adjusted for CO emission factors. After calculating $DM_{b,x}$ for each grid, we derive a regional average emission factor by dividing total CO emissions by total DM for each major BB region:

 $EF_{CO} = \sum_{b,x} CO / \sum_{b,x} DM$

These calculations enable us to discern the influence of LULC classification on BB emission inventories. For a specific biome type within a given region, we calculate EF by dividing the CO emissions of that particular biome classification by the sum of the value from each biome in the respective region:

$$EF_b = CO_b / \sum_b DM \tag{3}$$

(2)

248 where b represents one of the seven biome classifications in this study (Fig. S1).

Furthermore, for the two bottom-up inventories, we invert the fuel consumption for each vegetationbiome b within a given area:

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$$FC_b = DM_b/BA \tag{4}$$

Here, the DM corresponding to each biome in FINN1.5 and GFED4s is obtained using equation (1),and BA represents the total burned area derived from the emission inventory.

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255 **2.3 Quantitative statistical methods**

As described in section 2.1, fire detection is greatly affected by cloud/smoke obscuration in the bottom-up approach. For example, if there are clouds/smoke at high altitudes while fire occurs on the ground, the MCD14DL active fire detection product used in FINN1.5 may miss these fire points. In addition, the combustion that is too small in size and too low in temperature, cannot be effectively monitored due to the low brightness temperature contrast with the surrounding environment. In contrast, the burned area product (mainly MCD64A1) used by GFED4s determines the burning information based on the changes such as surface albedo, and is, therefore less affected by clouds/smoke. For inventories based on the top-down approach, the emission inventories also differ to a large extent due to the cloud/smoke obscuration, since QFED2.5 uses a "sequential method" to correct for missing FRPs during cloud/smoke obscuration, whereas VFEI0 does not. Thus, in this study, the symmetrical mean absolute percentage error (SMAPE) and Pearson's R are used to assess the difference in sensitivity to clouds/smoke between the two BB products based on the bottom-up (or top-down) approach. The specific algorithm is as follows:

269 $SMAPE = \frac{100\%}{n} \sum_{i=1}^{n} \frac{|X-Y|}{(|X|+|Y|)/2'}$

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$$R = \frac{\sum_{i=1}^{N} |(X - \bar{X}) \cdot (Y - \bar{Y})|}{\sqrt{\sum_{i=1}^{N} (X - \bar{X})^2 \cdot \sum_{i=1}^{N} (Y - \bar{Y})^2}},$$
(6)

(5)

where X and Y are fire detection data from two different datasets (e.g. burned area from FINN1.5 and GFED4s or FRP from VFEI0 and QFED2.5). We divided these fire detection data into three groups according to the cloud fractions less than 0.4, 0.4-0.7, and greater than 0.7, and the number n represents valid samples in different cloud fraction groups. SMAPE ranges from 0% to 200%, with smaller values indicating smaller differences, while Pearson's R ranges from 0 to 1, with smaller values implying less correlation.

In order to quantify the effect of cloud obscuration on BB datasets, we selected the most intensely 277 278 burning regions in BONA in July for this study. For consistency, we re-interpolated the fire detection data used in the four BB datasets, as well as the MODIS MCD06 cloud fraction data, to 279 280 the same horizontal resolution $(0.25^{\circ} \times 0.25^{\circ})$. Considering the continuity of combustion, we took every $5^{\circ} \times 5^{\circ}$ as a sample area in the northern U.S. to ensure that if a large burn occurred, the area 281 282 would be detected to some extent, avoiding errors due to differences between the inventories. At 283 the same time, we excluded the samples at the same time and location, where the emissions are all 284 zero. Finally, a total of 1888 samples were obtained for the burned area group, with 534, 541, and 285 813 samples for low (<0.4), medium (0.4-0.7), and high (>0.7) cloud fraction, respectively. A total of 1,682 samples were obtained for the FRP group, with 860, 390, and 432 samples under low, 286 medium, and high cloud fraction, respectively. It is worth noting that we use the average FRP of 287 288 MOD and MYD for QFED2.5 since the VFEI0 FRP is the average between day and nighttime 289 observations. Moreover, our approach cannot rule out the case of missing measurements when two 290 sets of BB inventories are both obscured by the cloud. However, the main goal of this paper is to explore the causes of uncertainties in emission inventories, the specific case of omission due to 291 292 cloud obscuration depends on the development of satellite detection technology and is not part of 293 the purpose of this study.

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295 **2.4 CESM2-CAM6 model**

296 The Community Earth System Model version 2.1 (CESM2) is a new generation of the coupled 297 climate/Earth system models developed by National Center for Atmospheric Research (NCAR). In this study, we used the global Community Atmosphere Model version 6 (CAM6) (Danabasoglu et 298 299 al., 2020). Gas-phase chemistry was represented by the Model for Ozone and Related chemical Tracers tropospheric chemistry (MOZART-T1, Emmons et al., 2020). The wet deposition of soluble 300 301 gaseous compounds in CAM6-Chem is based on the scheme of Neu and Prather (2012), which 302 describes the process of in-cloud cleaning and under-cloud cleaning. The formation of secondary 303 organic aerosols (SOA) is from a volatility basis set (VBS) approach developed by Tilmes (2019).

Properties and processes of aerosol species of black carbon (BC), primary organic aerosols (POA),
SOA, sulfate, dust, and sea salt are calculated by Modal Aerosol Module (MAM4) described by Liu
(2016). CAM6 uses a horizontal resolution of nominal 1° (1.25° × 0.9°, longitude by latitude) and
32 vertical levels from the surface to 2.26 hPa (~40 km).
In this study, four BB emission inventories (FINN1.5, GFED4s, QFED2.5, and VFEI0) are regridded to a horizontal resolution of 1.25° (longitude) × 0.9° (latitude), and then applied to the

- model. All simulations were performed for five years, while horizontal winds and temperature were
- 311 nudged toward the Modern-Era Retrospective analysis for Research and Applications, version 2
- 312 (MERRA-2) reanalysis data (GMAO, 2015) for every 6 h. Simulations are conducted for 2012-
- 2016, with the first year used for initialization and model spin-up. Daily BB emissions were applied
- in this study, whereas the vertical distribution of fire emissions was followed Freitas et al. (2006,
 2010). Anthropogenic and biogenic emissions in this study are from the Community Emissions Data
- 316 System (CEDS) and Model of Emissions of Gases and Aerosols from Nature version 2.1
- 317 (MEGANv2.1), respectively, at 2010 levels (Guenther et al., 2012; Hoesly et al., 2018).
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319 2.5 Measurement data

320 The Tropospheric Pollution Measurement Instrument (MOPITT) is aboard the Earth Observing 321 System (EOS)/Terra satellite launched by NASA (Warner, et al., 2001). MOPITT is the first instrument to observe the global concentration and currently provides column concentration and 322 323 volume mixing ratio of global carbon monoxide (CO) since 1999. We used MOPITT CO gridded monthly means (Near and Thermal Infrared Radiances) V009 (MOP03JM 9; NASA Langley 324 325 Atmospheric Science Data Center DAAC, retrieved from https://doi.org/10.5067/TERRA/MOPITT/MOP03JM.009), which has a horizontal resolution of 1° 326 327 \times 1°. It should be noted that to compare the CO column concentration simulated by CESM2-CAM6 328 with MOPITT CO, we calculated the simulated CO column concentrations by cumulative 329 integration from 900 hPa to 100 hPa isobaric height (Deeter et al., 2022). We also used the daily AOD (550 nm) and cloud fraction data from MODIS products MOD08 D3 (MODIS/Terra Aerosol 330 Cloud Water Vapor Ozone Daily L3; Platnick et al. 2015) and MCD06COSP (MODIS (Aqua/Terra) 331 Cloud Properties Level 3 daily, Webb et al., 2017), respectively. 332

The observations of AERONET (<u>http://AERONET.gsfc.nasa.gov/</u>; Holben et al., 1998) from 12 sites are used in this study. These AERONET stations were selected since they are close to BB source regions. As marked in Figure 1b, these sites include sites in BONA (Yellowknife_Aurora (62.5°N, 114.4°W), Pickle Lake (51.4°N, 90.2°W)), BOAS (Tiksi (71.6°N, 128.9°E), Yakutsk (61.7°N, 129.4°E)), SHAF (Namibe (15.2°S, 12.2°E), Mongu Inn (15.3°S, 23.1°E)), SHSA (Alta Floresta (9.9°S, 56.1°W), Rio Branco (9.9°S, 67.9°W)), EQAS (Palangkaraya (2.2°S, 113.9°E), Jambi (1.6°S, 103.6°E)), SEAS (Omkoi (17.8°N, 98.4°E), Ubon Ratchathani (15.2°N, 104.9°E)).

All observed AOD represent real atmospheric conditions and therefore, in addition to BB aerosols,
biogenic aerosols, anthropogenic aerosols, dust, and sea salts are also integrated in MODIS and
AERONET datasets.

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344 **3** Comparative analysis of emission inventories

CO and OC are the main species emitted from biomass burning (Westerling et al., 2010; van der
Werf et al., 2010b; Carter et al., 2020) but emissions vary widely. In this study, we compare the

differences in CO and OC emissions (representing gaseous and particulate pollutants, respectively)
in four BB inventories, and investigate in detail the key reasons for the differences in emission
inventories.

350 3.1 The contribution of dry matter and emission factors to the difference in CO 351 emission

352 The total global CO emissions from the four BB emission inventories selected for this study are in the range of 252-336 Tg, with GFED4s being the highest and FINN1.5 the lowest. To quantify 353 354 the differences in CO emissions among four datasets, we use the standard deviation (SD) to characterize the absolute difference, and the coefficient of variation (cv, calculated as the ratio of 355 SD to the mean) to characterize the relative differences (Fig. 2a). The larger the cv, the greater the 356 357 difference between emission inventories. We have ranked the major seven BB regions in the world according to the differences in CO emissions between the four sets of inventories, with the 358 359 differences being, in descending order, EQAS, BONA, SEAS, SHAF, NHAF, BOAS, and SHSA.

360 This study points to a high variability of different BB emission inventories in EQAS, which is 361 inconsistent with previous studies (Liu et al., 2020b; Pan et al., 2020). Previous studies mainly 362 focused on emission differences of particulate pollutants, such as BC and OC (Bian et al., 2007; 363 Paton-Walsh et al., 2012; Carter et al., 2020; Lin et al., 2020b; Pan et al., 2020), thus assuming that 364 the inventory differences in Equatorial Asia are smaller than those in Southern Hemispheric Africa 365 and Northern Hemispheric Africa. In contrast, this study analyzes the differences between 366 particulate and gaseous pollutant emissions separately when comparing the differences in BB 367 emission inventories. For example, GFED4s classify a large portion of EQAS land cover as peatland 368 (Kasischke and Bruhwiler, 2002; Stockwell et al., 2016; van der Werf et al., 2006, 2010a, 2010b, 369 2017) and suggest that this organic matter-rich soil emits a large amount of CO when burned. The 370 other three inventories either do not include peatland (FINN1.5 and QFED2.5) or only consider peatlands as a small fraction of the burned area in EQAS (VFEI0), thus estimating CO emissions 371 372 much smaller than GFED4s. In addition, the extent of peatland fires in EQAS increased significantly 373 during the strong El Niño event (Page et al., 2002). Considering that a strong El Niño event also occurred in 2015-2016, these increases in peatland fires further amplify the discrepancy between 374 375 GFED4s and other emission inventories on CO estimates.

As shown in Fig. 2, the distribution pattern of DM differences is very similar to that of CO emission differences, indicating that DM is the main reason for dominating the difference in the four emission inventories. In comparison, the difference in DM contributes 50-80% to the regional CO emission differences, and the comprehensive EFs contributes the remaining 20-50%. However, in EQAS, BONA, and BOAS, the contribution of comprehensive EFs to BB emission differences in four datasets is comparable to that of DM (Fig. 2). In the following sections, we will further analyze the main causes of the differences for DM and EFs.

383 3.2 Primary causes of DM inconsistency in the bottom-up inventories

To investigate the underlying causes of the differences in DM, we first compared DM between emission inventories produced by the bottom-up and up-down approaches. The difference in DM estimated by the top-down method is small, and the DM ratio of QFED2.5 to VFEI0 does not exceed two times in different regions. However, DM estimated by the bottom-up approach varied widely, with DM ratio as high as 4.7 in BONA for GFED4s and FINN1.5 during the 2013-2016 fire season.

Therefore, we need to focus on the main reasons for DM variance in emission inventories based onbottom-up approach.

391 According to Eq. (2), DM equals the product of the burned area, fuel load, and FB in the bottom-392 up inventories, with the product of the last two terms being fuel consumption. Fig. 3 compares the 393 burned area and fuel consumption of GFED4s and FINN1.5 emission inventories for the seven 394 largest BB regions. The ratio GFED4s/FINN1.5 represents the relative difference in burned area or 395 fuel consumption between the two emission inventories. In general, the difference in burned area 396 between the two inventories varies greatly with latitude, and the ratio of GFED4s to FINN1.5 397 fluctuates in the range of 0.28-1.94. In contrast, differences in fuel consumption between the two 398 inventories were more consistent, with GFED4s consistently having higher fuel consumption than 399 FINN1.5 in all regions except SEAS. In the next sections, we discuss the main reasons for the 400 differences in burned area and fuel consumption between the two datasets.

401 **3.2.1 Effect of land cover on burned area**

As shown in Fig. 3a, the differences in the burned area between the bottom-up emission 402 403 inventories are highly variable. At high latitudes, the burned area of GFED4s is significantly higher 404 than that of FINN1.5, especially in BONA, where the burned area of GFED4s is twice that of 405 FINN1.5. In contrast, the burned area of GFED4s in the equatorial region is much lower than that 406 of FINN1.5, and even 60% smaller in EQAS. This is a result of the difference in fire detection 407 between the two datasets. As shown in Table 1, FINN1.5 uses the MCD14 DL fire point product, 408 while GFED4s uses the hybrid burned area product, mainly using MCD64A1 combined with fire 409 point products MOD14A1/MYD14A1 to enhance the detection of small fires.

These two sets of products have their advantages in detection ability under different vegetation 410 411 type conditions. The hybrid burned area product detects burned areas over a period of time (up to 412 days), while the fire point product detects burned areas primarily in near real-time (Roy et al., 2008). 413 In addition, the burned area used in GFED4s (hybrid burned area product) is not affected by the 414 vegetation canopy when the leaf area index (LAI) is less than 5. Therefore, a higher burned area is estimated in GFED4s in BONA and BOAS than in FINN1.5. However, in areas with more broadleaf 415 416 forests and grasslands such as EQAS, SEAS, and SHSA (Fig. S2), the MCD14DL fire point product 417 used in FINN1.5 performed better in capturing understory fires that occurred in closed canopies 418 (Cochrane and Laurance, 2002; Cochrane, 2003; Alencar et al., 2005; Roy et al., 2008). It also has 419 an advantage in capturing sporadic and fragmented small fires in grasslands and agricultural fields 420 due to its high resolution (Liu et al., 2020b). Furthermore, FINN1.5 assumes that each detected fire 421 in the equatorial region will continue to burn for 2 days, and that the next day's fire will continue to 422 be half the size of the previous day (Table 1). Thus, the burned area of FINN1.5 in the tropical zone 423 is 2.6 times higher than that of GFED4s, which is consistent with previous studies (Wiedinmyer et 424 al., 2011; Pan et al., 2020). At the equator, the burned area in grassland/agricultural fields and forests 425 estimated by FINN1.5 is 1-3 and 4-6 times higher than in GFED4s, respectively (not shown).

It is worth noting that in Africa (NHAF and SHAF), although the dominant burnable vegetation
is grassland (Fig. S2), unlike the sporadic small fires that occur in grassland in the other five regions,
large continuous fires often occur in African Savannas (Liu et al., 2020b). Therefore, the hybrid
burned area product used in GFED4s is more effective in detecting all fire events occurring over
time, with 10-20% higher burned area than FINN1.5.

431 **3.2.2 Effect of cloud obscuration on burned area**

In addition to the vegetation, cloud occlusion can likewise bias the satellite detection of burned 432 433 area. Figure S3 shows the time series of AOD measured by satellite or ground-based data at the 434 Pickle Lack site of BONA from June to August 2013. In contrast to the high AOD values observed 435 for the AERONET network, MODIS AOD often in missing measurements when the MODIS cloud fraction is larger than 0.5. Furthermore, AERONET AOD varies dramatically over a short period, 436 437 suggesting that different detection principles (such as detecting fire points in near real-time during 438 satellite overpass time, or estimating the accumulation of burned area over time through changes in 439 surface albedo over multiple satellite overpass times) can significantly affect the burned area 440 product under high cloud fraction/smoke conditions (Paton-Walsh et al., 2012; Liu et al., 2020b; 441 Pan et al., 2020). Although some assumptions are made in FINN1.5 in the equatorial regions as 442 described above to improve the effect of cloud obscuration on burned area detection, these assumptions are not used for mid- and high-latitudes. GFED4s uses a hybrid burned area product 443 444 and is relatively unaffected by cloud obscuration. By fusing the MCD64A1 with MOD14A1/MYD14A1 products with multi-temporal satellite data, GFED4s is able to determine 445 the approximate date and extent of fires through post-fire ash deposition, vegetation migration, and 446 447 land surface changes (van der Werf et al., 2017; Boschetti et al., 2015, 2019).

448 To quantitatively assess the impact of cloud obscuration on different emission inventory estimates, 449 we perform analyses in areas with high cloud fraction (Fig. S4), intense biomass burning, and 450 unaffected by the smoothing hypothesis used in FINN1.5. We selected the regions of North America 451 with the most intense biomass burning (Alberta and Saskatchewan, Canada, 50°-70°E, 100°-130°W, 452 Fig. S5), and analyzed the relationship between the burned area and cloud fraction for bottom-up 453 inventories during July from 2013 to 2016 (Fig. S6). As shown in Fig. 4, with the increase in cloud 454 fraction, the SMAPE of the two bottom-up emission inventories increases from 150% to 180%, 455 while the Pearson correlation declines from 0.85 to around 0.75. These results demonstrate that the 456 uncertainty in the burned area for two bottom-up emission inventories increases by $\sim 20\%$ during 457 high cloud fraction compared to low cloud fraction conditions.

458 **3.2.3** Causes of Fuel Consumption differences

459 Fuel consumption is another factor that affects DM differences between two BB emission 460 inventories. As shown in Fig. 3b, the fuel consumption of GFED4s is 30-75% higher than that of FINN1.5 in almost all BB areas except SEAS. The difference in fuel consumption between the two 461 emission inventories is larger in the tropics than in the high latitudes. As shown in Fig. 5, at high 462 463 latitudes (e.g., BONA and BOAS), and in the equatorial region (such as EQAS), relatively high fuel 464 consumption comes from peatlands in GFED4s. According to previous studies, peatlands, a type of soil rich in organic matter, store large amounts of carbon underground (van der Werf et al., 2010b, 465 466 2017; Gibson et al., 2018; Kiely et al., 2021; Vetrita et al., 2021), and emit large amounts of CO 467 when burned. Peatlands contribute 30-60% of the total fuel consumption in BONA, BOAS, and 468 EQAS (Fig. 5a-c).

Besides peatlands, GFED4s tends to have higher fuel consumption than FINN1.5 due to forest
contributions. Forests (including tropical, temperate, and boreal forests) account for more than 50%
of the fuel consumption in all burning regions except EQAS, where peatlands dominate the fuel
consumption. Moreover, forest fuel consumption in GFED4s is generally much higher than in

473 FINN1.5 except in BOAS and SEAS (Fig. 5). Since fuel consumption is equal to the product of fuel 474 load and FB (the percentage of specific plants that can be adequately burned, Eq. 2), different 475 vegetation classifications may be responsible for large differences in fuel consumption between 476 emission inventories. For example, for woody vegetation such as forests, GFED4s assumes a range 477 of FB between 40-60% for temperate and tropical forests and 20-40% for boreal forests, while 478 FINN1.5 assumes that all woody vegetation burns no more than 30% (van der Werf et al., 2010; Wiedinmyer et al., 2011). Thus, in terms of FB alone, the forest fuel consumption of GFED4s is 479 480 therefore 0.67-1.3 times greater than that of FINN1.5, which is one of the main reasons for the 481 difference in fuel consumption.

482 **3.3** Primary causes of DM inconsistency in the top-down approach

We also analyze the causes of the difference in DM between BB emission inventories estimated 483 484 by the top-down method. According to Eq. (3), it is evident that the empirical factor and the radiative 485 energy of the fire are the key factors that cause the discrepancy in the top-down emission inventories. 486 The QFED2.5 and VFEI0 inventories we have chosen use different satellites for the fire detection 487 products. For example, for the fire radiative power product, QFED2.5 is based on the Moderate 488 Resolution Imaging Spectroradiometer (MODIS) inversion of the NASA Terra and Aqua combined satellites, while VFEI0 is based on the Visible Infrared Imaging Radiometer (VIIRS) inversion of 489 490 the combined polar-orbiting satellites Suomi NPP and NOAA-20, although the algorithms are 491 similar. However, there are systematic deviations due to different satellites, specific tests and 492 metadata, and resolutions. The VIIRS 375 m fire product used by VFEI0 has a finer resolution and 493 is more advantageous for small fire spot detection than other coarser resolution (1 km) fire spot 494 detection products. The FRP density used in VFEI0 is much higher than that of QFED2.5 due to the 495 fine horizontal resolution.

496 The estimations of FRP and DM are strongly influenced by the horizontal resolution of satellite 497 products. For example, in the BONA region during July (the month with the most intense burning at the position of 50°-70°N, 100°-130°W), the total QFED FRP (average FRP measured by MOD 498 and MYD) is 1.5 times higher than VFEI0 (Fig. S7). Additionally, the differing α values between 499 500 QFED2.5 and VFEI0 in BONA can potentially result in higher DM in QFED2.5 compared to VFEI0 by a factor of 1.3-3.8. However, the actual DM in the QFED2.5 inventory is 30% lower than in 501 502 VFEI0. The relatively high FRP density used in VFEI0 (Fig. S8) results in a higher DM than in 503 QFED2.5 due to its superior horizontal resolution, enabling the precise delineation of fire areas. It is important to note that while the empirical factor also influences the amount of DM, its impact 504 505 should not be as significant as the difference caused by the horizontal resolution of satellite products 506 (Kaiser et al., 2012; Darmenov et al., 2015; Ferrada et al. 2022).

507 Previous studies have shown that cloud occlusion also causes bias in FRP detection (Liu et al., 2020b). We also take BONA as a pilot region to analyze the influence of cloud fraction on FRP in 508 509 QFED2.5 and VFEI0. According to Fig. 5c-d, the SMAPE of the two emission inventories rises as the cloud fraction increases, and the Pearson correlation is noticeably low under the maximum cloud 510 fraction. While QFED2.5 uses the "sequential approach" (section 2.1) to correct for the missing 511 512 FRP in cloud-obscured fires, this correction is not considered in VFEI0. Therefore, although the two 513 top-down emission inventories use similar algorithms, significant bias occurs under high cloud fraction conditions, with QFED2.5 estimating DM much higher than VFEI0. 514

515 **3.4 Primary causes of EF inconsistencies**

Although DM differences dominate the inconsistencies of CO emissions across major BB regions, 516 517 the contribution of EFs is still not negligible in some regions. For example, in EQAS, BONA, and 518 BOAS, the contribution of EFs is up to 50%, which is comparable to that of DM. The comprehensive 519 EFs of GFED4s are higher in BONA, BOAS, and EQAS regions than in other inventories, with vegetation classification being one of the most important factors (Fig. 6). For example, in EQAS at 520 521 low latitudes, peatlands in GFED4s account for 65% of the regional comprehensive EF. In contrast to GFED4s, FINN1.5, and QFED2.5 do not consider this organic matter-rich land as a source of 522 523 burning, and they classify this category of land cover type as savanna or grass. The CO emission factor for peatlands is four times higher than the CO emission factor for savanna or grass (Table 2), 524 525 ultimately making the comprehensive EF for GFED4s 60-70% higher than that of the other three 526 datasets. It is worth noting that although the classification of Peatland exists in VFEI0 (Ferrada et 527 al., 2022), due to differences in terrestrial ecological divisions (Olson et al., 2001; 528 http://www.worldwildlife.org/science/data/item1875.html), peatlands identification areas are much 529 smaller than GFED4s inventory. Therefore CO emissions from peatlands in GFED4s are much higher than in the VFEI0 inventory (Figure 3-9a; Ferrada et al., 2022). 530

531 In both BONA and BOAS, we find that the comprehensive EFs in the four datasets are ranked as follows: GFED4s>FINN1.5>QFED2.5>VFEI0, where the EF of GFED4s is about 1.5 times higher 532 than that of VFEI0. Unlike the low-latitude regions, the classification of forests in different emission 533 534 inventories is the main reason for the difference in comprehensive EF in high-latitude regions. At 535 high latitudes (50° - 70°N), GFED4s, QFED2.5, and FINN1.5 identify more forests than VFEI0 536 (Fig. S1) because the former three classify some shrubs (e.g., closed shrublands and woody savanna) 537 as forests, while the latter classify them as grassland. Forests contribute to 70% or more of the 538 comprehensive EFs at high latitudes in the first three emission inventories, but only 8% to the 539 comprehensive EF in VFEI0. The remaining gap in the absolute contribution of forests is caused by 540 the difference in the selected emission factors and the horizontal resolution of the satellite products.

541 **3.5** Contribution of DM and EFs to differences in OC emissions

542 The above analysis completes the comparison of gaseous pollutant CO among different emission 543 inventories. In this section, we will take OC as an example to compare the emission differences of 544 particulate pollutants. As shown in Fig. 7, the global OC emissions of four datasets range from 14.9 to 42.9Tg, with the highest emissions from QFED2.5, which is consistent with previous studies 545 (Carter et al., 2020; Pan et al., 2020). According to the statistical method in section 3.1, we 546 547 quantified the magnitude of OC emission differences between regions and ranked them as follows: 548 BONA>BOAS>NHAF>SHAF>SEAS>SHSA>EQAS. Compared to the CO emission differences (Fig. 2), the difference in OC emissions becomes larger for BOAS and smaller for low latitude 549 regions of SEAS and EQAS. Since DM should be consistent in the same emission inventories for a 550 551 given time and area, the magnitude of emissions for different species depends on changes in 552 emission factors. Considering that the emission factors of aerosol-related emission species such as 553 OC, BC, NH₃, SO₂, and PM_{2.5} have been corrected based on the satellite retrieved AOD of the QFED2.5 emission inventory (Table 2), the EFs of OC in QFED2.5 are much higher than that of the 554 555 other three emission inventories (Fig. 7b). As a result, the OC EFs in the QFED2.5 emission 556 inventory were enlarged by a factor of 1.8-4.5 times through the correction of BOAS, SEAS and

557 EQAS (Table 2). In contrast, the other three emission inventories were not corrected for OC EFs.

- 558 Unlike the CO EFs, the OC EFs of GFED4s in equatorial regions are largely consistent with the
- 559 FINN1.5 and VFEI0 emission inventories. Although burning organic matter-rich soil substrates is
- 560 generally thought to release large amounts of CO, their ability to release OC is similar to that of
- vegetation such as shrubs and some forests. Thus, despite CO emissions bias in EQAS being largely

affected by peatlands, differences in OC emissions among the four inventories are not significant.

Compared with Pan et al. (2020), it is obvious that the top-down approach will not lead to an increase in emission deviation of the particulate-phase species. The correction of EFs, however, is the root cause of the increased bias in OC emissions. Pan et al. (2020) reported that QFED2.5 and FEER1.0 had the highest global OC emissions, while GFAS1.2 had much lower OC emissions. In this study, the largest OC emission also appears in QFED2.5, but the global total OC emissions of the recently released VFEI0 are relatively low.

569 4 Model evaluation based on emission inventories application

570 4.1 Comparison of simulations with MOPITT CO

571 One of the main goals of this study is to provide a confidence assessment of the BB emission 572 inventories by comparing model simulations with observations. A comparison between model 573 simulations using different emission inventories and ground-based/satellite-retrieved data for the 574 respective fire seasons (Table 3) of the main BB regions is explored below. In this study, we 575 compared the model results with measurements from two perspectives: the spatial distribution of 576 BB pollutants, and the time-varying characteristics of BB pollutants.

577 Figure 8 depicts the spatial distribution of CO column burdens in SHSA and SHAF during the 578 fire seasons. In SHSA, the simulated CO column burdens using different emission inventories are all consistent with the spatial distribution pattern of MOPITT CO column burden, with the peak 579 value located in the Amazon rainforest. However, the central value of MOPITT CO column burden 580 is as high as 2.8×10^{18} molecules cm⁻², which is slightly higher than the simulated results. Among 581 the four sets of emission inventories, the peak amplitude and spatial distribution of simulated CO 582 583 column burdens are closest to the satellite-retrieved data after applying the GFED4s and VFEI0. In 584 SHAF, however, the model underestimated the peak CO column burden after applying all emission 585 inventories except VFEI0.

586 In addition to SHSA and SHAF, a comparison of regionally averaged CO column burdens 587 between our simulations and MOPITT CO in major BB regions is also shown in Table 3. In the Northern Hemisphere, our simulations are significantly underestimated compared to MOPITT CO, 588 589 while those in the Southern Hemisphere are consistent with satellite retrievals. Surprisingly, the 590 simulated spatial distributions and magnitudes of CO in the Southern Hemisphere using the recently released VFEI0 agree very well with observations. In contrast, the underestimation of CO 591 592 concentrations in the Northern Hemisphere is partly due to uncertainty in anthropogenic emissions, 593 as we assume anthropogenic emissions at 2010 levels, which are lower than those during the 2013-594 2016 period.

Note that simulated CO concentrations are 30-40% lower than MOPITT CO at high latitudes.
Besides the impact of emission inventories, there are also large uncertainties in satellite-retrieved
CO concentrations (Lin et al., 2020a; Pan et al., 2020). In addition, OH loss, long-range transport,
and photochemical reactions involved in the CESM2-CAM6 model simulations also lead to

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uncertainties in simulated CO. For example, MOZART-4x contains an additional OH oxidation
pathway for CO, which may lead to lower CO concentrations (Lamarque et al., 2012; He and Zhang,
2014; Barré et al., 2015; Brown-Steiner et al., 2018; Emmons et al., 2020). In comparison, the
simulated CO by using GFED4s is closest to the MOPITT CO value in terms of spatial distribution
and peak magnitude at high latitudes in the Northern Hemisphere, which is superior to other
emission inventories.

605 4.2 Comparison of simulations with MODIS AOD

606 We compared MODIS-derived aerosol optical depth (AOD) data with simulated AOD in major 607 BB areas. Figure 9 shows the spatial distribution of AOD in SHSA and SHAF during their fire 608 seasons. The simulated AOD is significantly higher than the MODIS AOD in SHSA. Note that 609 primary organic aerosols (POA) associated with BB account for only 15-23% of the total AOD in 610 Amazon, while secondary organic aerosols (SOA) account for approximately 50% of the total AOD. Furthermore, overestimation of simulated AOD occurs throughout the year, not just during the fire 611 612 season. Considering the high biogenic emissions in this region, the overestimation of AOD could 613 be attributed to the formation of biogenic SOA (He et al., 2015; Tilmes et al., 2019). In SHAF, the 614 spatial distribution and magnitude of simulated AOD using GFED4s and VFEI0 are close to those of the MODIS AOD. In comparison, our results show that AOD is significantly underestimated 615 616 using FINN1.5, but largely overestimated using QFED2.5.

617 Table 4 shows the mean values of model-simulated AOD and satellite measurements for each 618 region during its fire season. The influence of the BB emission inventory has little effect on the 619 simulated AOD value in the Southern Hemisphere, and the regional average AOD deviation is within 20%. In contrast, the average deviation of simulated AOD driven by four BB inventories can 620 621 be as high as 40% in the high latitudes of the Northern Hemisphere. Comparatively, GFED4s and 622 QFED2.5 are more suited for high latitudes in the northern hemisphere, whereas the VFEI0 is most 623 suitable for the southern hemisphere for AOD simulations. In Africa, QFED2.5 is not recommended due to its considerable overestimation. 624

625 4.3 Comparison of simulations with ground-based measurements

In the above sections, we merely discussed the spatial distribution and the magnitude of pollutants during fire seasons. To further analyze whether each dataset can effectively capture the instantaneous combustion of BB, we compared the value of simulated daily AOD with that of ground-based observation (Fig. 10). To be more representative, we selected stations in each BB region with a large amount of data during fire season, allowing a comprehensive assessment of the global BB emission inventories. The specific locations of the selected 12 AERONET sites are shown as red triangles in Fig. 1b.

At EQAS sites such as Palangkaraya and Jambi, the observed AOD from September to November 2014/2015 is generally higher than 1, with peaks exceeding 5, reflecting the intense BB events (Fig. 10a-b). Only simulations using GFED4s are consistent with observed AOD during strong BB events, with a slight underestimation of 33-38%, while none of the other simulations could capture the BB process. Considering the significant contribution of peatlands to BB emissions in EQAS in GFED4s, our results suggest that it is important to include the burning of organic matter-rich soils in BB emission inventories. At SEAS sites such as Omkoi and Ubon Ratchathani, the peak AOD occurs 640 from February to April at a value of about 2, and all simulations applying the four emission 641 inventories capture the observed changes in AOD (Fig 10c-d). However, due to the uncertainty of 642 anthropogenic emissions, the simulated AOD is usually smaller than the actual observed value in 643 EQAS. Note that simulations using QFED2.5 are most consistent with observed AOD during intense 644 biomass burning events.

645 At the Namibe station of SHAF (Fig. 10e), the simulated AOD agrees best with the measured 646 results after using FINN1.5 and GFED2.5, with NMB values within $\pm 8\%$, indicating these two 647 emission inventories can characterize the day-to-day variability of the intense BB process. However, 648 Namibe is located downwind of the dust source, and dust aerosols contribute more than 50% to the 649 total AOD in this area. To better evaluate the performance of the four BB emission inventories in 650 SHAF, we chose another site, Mongu Inn, located in the interior of Southern Hemispheric Africa, 651 where dust and sea salt accounted for 20-30% of the total AOD. At Mongu Inn, all simulations underestimate AOD by 46-71%, and only QFED2.5 and VFEI0 emission inventories can capture a 652 few peaks during intense biomass burning events (Fig. 10f). In SHSA, while Figures 9 and 10h 653 654 show an overall overestimation of simulated AOD compared to MODIS AOD, at the Brazilian Alta 655 Floresta site east of the Amazon, simulated AOD agrees very well with the ground-based observations (Fig. 10g). In general, the simulations using the VFEI0 emission inventory for the 656 657 Southern Hemisphere are close to the measurements.

At high latitudes, simulations driven by GFED4s and QFED2.5 better capture the observed peak
AOD, with regional NMB values of less than 40% (Fig. 10i-l), suggesting that these two simulations
can reproduce the intense BB process. In contrast, FINN1.5 and VFEI0 are obviously not suitable
for describing the BB process in these sites, and the simulated AOD is underestimated by 60-80%.

662 **5** Conclusion and Discussion

663 In this study, we examine four commonly used BB emission inventories (two bottom-up 664 inventories (GFED4s and FINN1.5) and two top-down inventories (QFED2.5 and VFEI0)) to better understand the uncertainties associated with BB emissions. We analyze variations in CO and OC 665 666 emissions across seven major BB regions worldwide from 2013 to 2016. We explore the differences 667 between gaseous and particulate emission inventories, quantifying the impact of vegetation 668 classification, cloud cover, and emission factors on inventory bias. Additionally, we apply these 669 inventories to the global model CESM2-CAM6 to assess the model performance in simulating 670 pollutants against satellite and ground-based observations.

671 The total global CO emissions exhibit significant variability among the four inventories, with 672 annual averages ranging from 252 to 336 Tg, and a maximum deviation rate exceeding 30%. In 673 certain regions such as BONA, changes in CO emissions are even larger, GFED4s emits 5.8 times 674 more CO than FINN1.5. DM is identified as the primary contributor to variance among BB emission inventories, accounting for 50-80% of regional bias, while comprehensive EFs contribute the 675 676 remaining 20-50%. Interestingly, the contributions of DM and comprehensive EFs to emission 677 inventory differences are comparable across equatorial regions and Northern Hemisphere high 678 latitudes.

The uncertainty in DM arises from underlying fuel consumption and burned area, linked to vegetation classification, fire detection product algorithm, and cloud/smoke masking. Vegetation classification significantly impacts fuel loading and the Fraction of Biomass burned, with discrepancies contributing to biases in fuel consumption. In regions at both low and high latitudes

(except Southeast Asia), FINN1.5 exhibits a fuel consumption term that is less than 50% of
GFED4s, with the vegetation classification methodology contributing primarily to this bias.
Different fire detection products introduce bias in estimated burned area, affecting uncertainty in
DM. Satellite transit/cloud obscuration influences DM by affecting burned area/fire radiative energy.
Cloud cover at high latitudes substantially impacts emission uncertainty, with bias increasing by 20%
in July in BONA under higher cloud fraction.

We extend our analysis to particulate pollutants, using OC emissions as an example. Global average annual OC emissions vary widely among the four inventories, ranging from 14.9 to 42.9 Tg, demonstrating greater variability than gaseous species like CO. BB OC emissions exhibit large variability at high latitudes in the Northern Hemisphere, with QFED2.5 adjusting emission factors based on satellite aerosol optical thickness (AOD) to enhance particulate matter emissions.

694 Applying four BB emission inventories to CESM2-CAM6, we compare model-simulated CO 695 column concentrations with the MOPITT satellite inversion CO column concentrations. According 696 to our simulations, CO simulated using GFED4s is closest to satellite observations in almost all 697 regions except southern Asia and Africa. We also compared model results with AOD retrieved from 698 MODIS satellites or measured by AERONET. Simulated AOD at high northern latitudes is often 699 underestimated when using current mainstream BB emission inventories. For example, the 700 simulated regional average AOD is 8-46% lower than MODIS in North America. Unlike the high 701 latitudes, the simulated AOD is significantly overestimated at the equator, and the regional average 702 AOD simulated by the model in Northern Hemispheric Africa is 66-91% higher than MODIS. In 703 addition, comparing model simulated AOD with AERONET ground-based observations, we find 704 that GFED4s performs best in EQAS for daily variability during intense burning. In SEAS, although 705 FINN1.5 can better represent the magnitude of the overall OC emissions in the BB season, QFED2.5 706 can capture the day-to-day variation characteristics of intense combustion. In the Southern 707 Hemisphere, the latest VFEI0 emission inventory performs well, and the simulated AOD is able to 708 capture the BB processes.

709 Our study assesses the global applicability of BB emission inventories and has some implications 710 for future studies. Overall, GFED4s and QFED2.5 inventories for the northern high latitudes capture 711 the magnitude and daily variation of OC emitted throughout the BB season. These two emission 712 inventories outperformed the others when applied to studies of interactions between BB aerosol and weather/climate. In the Southern Hemisphere, the spatial distribution and daily variation 713 714 characteristics of CO and AOD simulated by the model are closest to the observed values when the 715 latest VFEI0 emission inventory is applied. For the equator, the situation is more complicated, and 716 we recommend combining emission inventories according to the research objectives. For example, 717 GFED4s performs best in day-to-day changes during intense burning in equatorial Asia. In 718 Southeast Asia, combining OC magnitude in FINN1.5 and daily variation in QFED2.5 is the optimal 719 choice.

720 It is worth noting that emission factors (as listed in Table 2) significantly contribute to the 721 differences in BB emissions. However, actual emission factors vary widely depending on the 722 different states of combustion (Pokhrel et al., 2021). Further study is needed to understand the 723 impact of combustion efficiency on the BB EFs and optimize them.

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Data availability. The biomass burning emission datasets used in this work are available from
 http://www.globalfifiredata.org (GFED4s), https://www.acom.ucar.edu/Data/fire/ (FINN1.5),

727 https://portal.nccs.nasa.gov/datashare/iesa/aerosol/emissions/QFED/v2.5r1/ (QFED2.5), and http://bio.cgrer.uiowa.edu/VFEI/DOWNLOAD/ (VFEI0). AOD and cloud fraction from MODIS 728 729 dataset can be obtained from https://ladsweb.modaps.eosdis.nasa.gov/search/. MOPITT CO can be 730 obtained from https://doi.org/10.5067/TERRA/MOPITT/MOP03JM.009. AERONET AOD is 731 avaiable from https://aeronet.gsfc.nasa.gov/new web/download all v3 aod.html. The Modern-Era 732 Retrospective analysis for Research and Applications, version 2 (MERRA-2) reanalysis data is available from https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/. All data analyzed during the 733 734 current study are included in this published article and its supplementary information. Raw model 735 simulations are available from the corresponding author on reasonable request.

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Author contributions. S. L. and A. D. designed the research, W. H. and S. L. conducted the data
analysis and model simulations, W. H. and S. L. took the lead in writing the manuscript, with
contributions from all authors.

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742 *Competing interests.* The authors declare that they have no conflict of interest.

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1034	Cable 1. Brief introduction of four BB inventories	
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	"Bo	"Bottom-up" "Top-down"		vn"
	FINN1.5	GFED4s	QFED2.5	VFEI0
Temporal range	2002- (NRT) ^a	1997-2022 ^b	2000- (NRT) ^a	2012- (NRT) ^a
Spatio-temporal resolution	1km, daily	0.25°, monthly (daily fraction)	0.1°, daily (0.25° × 0.375°, NRT ^a)	500m, daily
Primary satellite fire input	MCD14DL C5 active fire	MCD64A1 C5.1 burned area	MOD14/MYD14 C6 FRP (1km)	VNP14IMG FRP (1km)
	area (1km)	(500m)		
Statistical	Smooth assumption	Small fire boost	Cloud-gap adjusted FRP density	
boosts/Adjustion	in tropics °	(MOD14A1/MYD14A1)		
Primary land use/land	MCD12Q1 (IGBP), 2005	MCD12Q1 (UMD), 2001-2012	IGBP-INPE	MCD12C1(IGBP) +
cover (LULC)				The Köppen Climate
				Classification
Peatland fire	×	Olson et al. (2001)	×	Ferrada et al. (2022)
Conversion to dry matter	Hoelzemann et al. (2004)	CASA biogeochemical model	QFED FRP vs GFED2 dry matter	VFEI FRP vs GFED3.1
		(van der Werf et al., 2010)	global calibration	matter global calibration
Emission factors	Akagi et al. (2011),	Akagi et al. (2011) + updates	Andreae and Merlet (2001),	Akagi et al. (2019)
	Andreae and Merlet	from Andreae et al. (2013)	Akagi et al. (2011) ^d	
	(2001)			
Speciation	41 species	27 species	17 species	46 species
References	Wiedinmyer et al. (2011)	van der Werf et al. (2017)	Darmenov and da Silva (2015)	Ferrada et al. (2022)

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1043 Table 2. CO and OC emission factors used in the four biomass burning emission inventories.

		Emission	factors across i	nventories and	l vegetation types (g species per	kg dry matter)	
Types		CC)				OC	
	FINN1.5	GFED4s	QFED2.5	VFEI0	FINN1.5	GFED4s	QFED2.5	VFEI0
Temperate forest	108 ^{Ak}	88 ^{Ak}	107 ^{am}	113 ^{An}	6.97 ^{ar}	9.6 ^{AM}	41.09*	10.9 ^{An}
Boreal forest	118 ^{Ak}	127 ^{Ak}	107 ^{AM}	121 ^{An}	7.31 ^{Mc}	9.6 ^{AM}	41.09*	5.9. ^{An}
Savanna and Grass, shrub	59 ^{Ak} /68 ^{Ak}	63 ^{Ak}	65 ^{am}	69 ^{An}	2.6 ^{Ak} /6.61 ^{Mc}	2.62 ^{Ak}	6.12*	3 ^{An}
Tropical forest	92 ^{Ak}	93 ^{Ak}	104 ^{AM}	104^{An}	4.77 ^{Ak}	4.71 ^{Ak}	13*	4.4 ^{An}
Agricultural	111 ^{Ak}	102 ^{Ak}	/	76 ^{An}	3.3 ^{AM}	2.3 ^{Ak}	/	4.9 ^{An}
Peatlands	/	210#	/	260 ^{An}	/	6.02#	/	14.2 ^{An}

1044 Ak: Akagi et al. (2011); AM: Andreae and Merlet (2001); An: Andreae (2019); AR: Andreae and Rosenfeld (2008); Mc: McMeeking et al. (2009)

1045 *: QFED2.5 PM-related emission factors are obtained by multiplying the base EF multiplied by its biome-specific enhancement factor

1046 #: Emission factors for peatland is the average of lab measurements of Yokelson et al. (1997) and Christian et al. (2003)

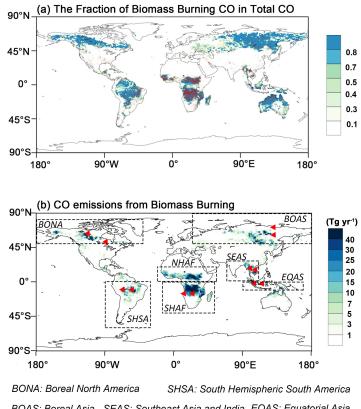
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Table 3. Comparison of CESM-CAM6 simulated CO column averages and satellite retrieved CO
column averages during the fire season.

Regions		Satellite		CESM	CESM2-CAM6	
	Fire-	MOPITT	FINN1.5	GFED4s	QFED2.5	VFEI0
	Season					
EQAS	JanApr.	1.88	1.66	1.69	1.61	1.47
BONA	AprAug.	2.03	1.29	1.47	1.30	1.32
SEAS	FebApr.	2.40	2.10	1.94	1.89	1.95
SHAF	MayNov.	2.31	1.75	2.04	1.99	2.19
NHAF	JanMay.	2.66	1.96	2.02	2.05	2.10
BOAS	MarNov.	2.05	1.31	1.42	1.33	1.34
SHSA	JulyDec.	1.77	1.75	1.80	1.76	1.80

	Satellite		CESM2-CAM6					
Regions	MODIS	FINN1.5	GFED4s	QFED2.5	VFEI0			
EQAS	0.23	0.22	0.25	0.23	0.21			
BONA	0.13	0.07	0.12	0.11	0.07			
SEAS	0.30	0.35	0.30	0.36	0.30			
SHAF	0.33	0.31	0.37	0.53	0.40			
NHAF	0.32	0.53	0.54	0.61	0.55			
BOAS	0.15	0.11	0.13	0.16	0.11			
SHSA	0.14	0.30	0.31	0.34	0.29			

1053 Table 4. Same as Table 3 but for AOD



BOAS: Boreal Asia SEAS: Southeast Asia and India EQAS: Equatorial Asia NHAF: Northern Hemispheric Africa SHAF: Southern Hemispheric Africa

1059 Figure 1. (a) The fraction of BB CO emissions to the sum of anthropogenic and BB CO emissions 1060 (CO BB/CO Total) during 2013-2016 and (b) the spatial distribution of CO emissions (FINN1.5 was used as an example). The red dots in Fig. 1(a) are the fire points from the MCD14DL satellite product. 1061 1062 In Fig. 1(b), seven regions with high BB emissions taken from those applied by van der Werf et al. (2006, 1063 2010) are marked with black boxes, and the red triangles represent 12 AERONET stations. In this study, seven major BB regions includes Boreal North America (BONA), Boreal Asia (BOAS), Southeast Asia 1064 (SEAS), Equatorial Asia (EQAS), North Hemisphere Africa (NHAF), South Hemisphere Africa (SHAF), 1065 and South Hemisphere South America (SHSA). 1066

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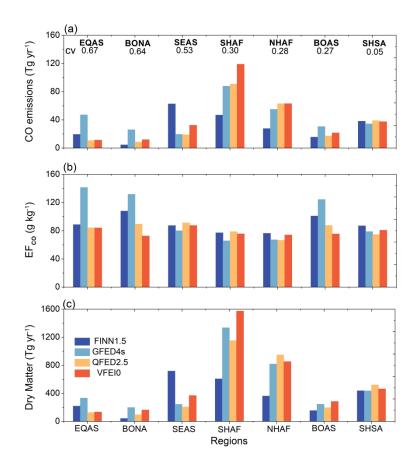
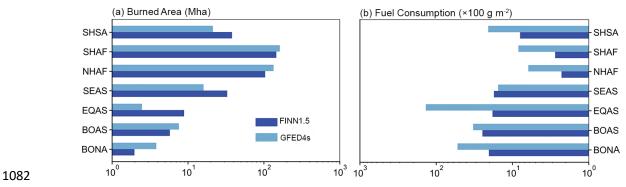


Figure 2. (a) Average annual CO emissions of four biomass burning emission inventories across seven
major BB regions during 2013-2016. The cv, defined as the ratio of the standard deviation to the mean,
is the coefficient of variation among the emissions of four datasets. (b) and (c) are the same as (a), but
for the emission factor of CO (EF_{CO}) and Dry Matter.

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1083 Figure 3. Annual burned area (a) and fuel consumption (b) of two bottom-up datasets (FINN1.5 and

1084 GFED4s) across seven regions from 2013 to 2016.

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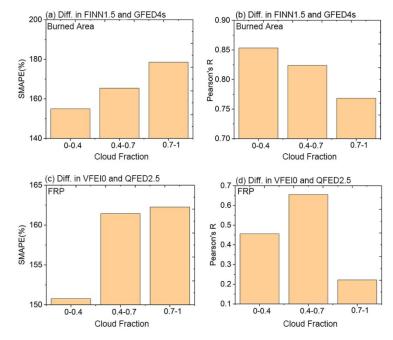
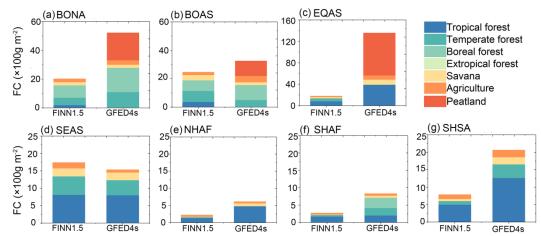
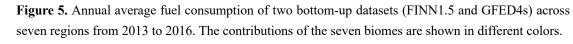


Figure 4. The differences in (a-b) burned areas and (c-d) total FRP detected by two inventories under
different cloud fraction in a pilot region of BONA. These differences are quantified by two indicators:
SMAPE and Pearson's R. Could fraction data is calculated from MODIS product MCD06COSP.

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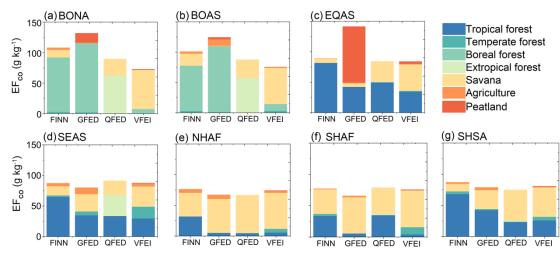


Figure 6. Regional comprehensive emission factors for four datasets (FINN1.5, GFED4s, QFED2.5,
and VFEI0) in seven regions from 2013 to 2016. The contributions of the seven biomes are shown in

- different colors.

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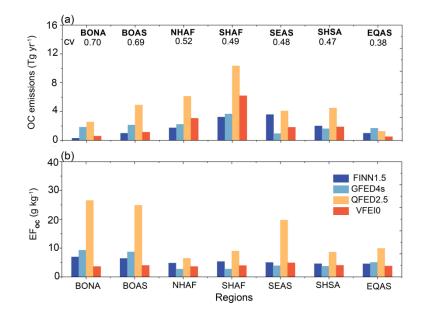


Figure 7. (a) Average annual OC emissions of four biomass burning emissions inventories across seven major BB regions during 2013-2016. The cv, defined as the ratio of the standard deviation to the mean, is the coefficient of variation among the emissions of four datasets. (b) is the same as (a) but for the emission factor of OC (EF_{oc}).

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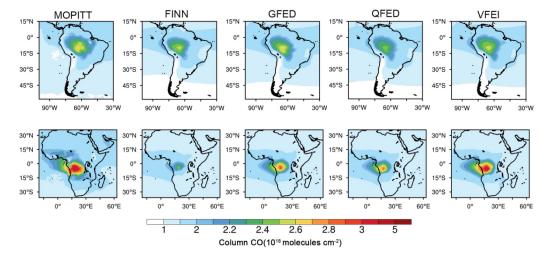


Figure 8. Spatial distribution of CO column burdens from MOPITT and CESM2-CAM6 simulations
during the fire season (Table 3). The text above each plot identifies the name of the satellite inversion
dataset or emission inventory dataset applied by the model, namely FINN1.5, GFED4s, QFED2.5, and
VFEI0.

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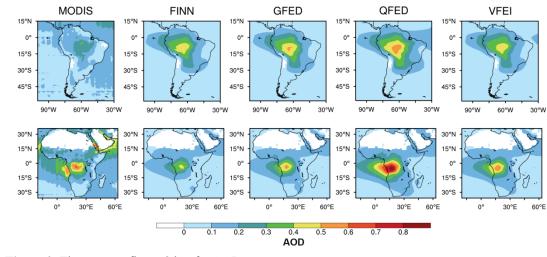


Figure 9. The same as figure 8 but for AOD.

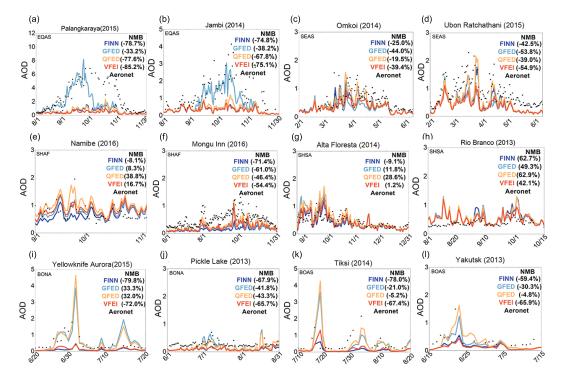




Figure 10. Comparison between AOD simulated by CESM2-CAM6 using the four datasets (FINN1.5,
GFED4s, QFED2.5, and VFEI0) and AERONET ground-based observations during fire seasons. These
AERONET sites are: (a) Palangkaraya (2.2°S, 113.9°E), (b) Jambi (1.6°S, 103.6°E), (c) Omkoi (17.8°N,
98.4°E), (d) Ubon Ratchathani (15.2°N, 104.9°E), (e) Namibe (15.2°S, 12.2°E), (f) Mongu Inn (15.3°S,
23.1°E), (g) Alta Floresta (9.9°S, 56.1°W), (h) Rio Branco (9.9°S, 67.9°W), (i) Yellowknife_Aurora
(62.5°N, 114.4°W), (j) Pickle Lake (51.4°N, 90.2°W), (k) Tiksi (71.6°N, 128.9°E), (l) Yakutsk (61.7°N,
129.4°E).