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# The export of African mineral dust across the Atlantic and its impact over the Amazon Basin

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## Abstract

The Amazon Basin is frequently influenced by the trans-Atlantic transport of African dust plumes during ~~the~~ its wet season, ~~(January – April)~~, which not only interrupts the near-pristine atmospheric condition in that season, but also provides nutrient inputs into the Amazon rainforest ~~associated with dust deposition. The factors controlling the long-range transport (LRT) of African dust towards the Amazon Basin and consequently the overall impact of African dust over the Amazon Basin are not yet well understood upon deposition.~~ In this study, we use the chemical transport model GEOS-Chem to investigate the impact of the export of African mineral dust ~~upon~~ to the Amazon Basin during the period of 2013 – 2017, constrained by multiple datasets obtained from AERONET, MODIS, as well as Cayenne site and the Amazon Tall Tower Observatory (ATTO) site in the Amazon Basin. With optimized particle mass size distribution (PMSD), ~~of dust aerosols,~~ the model well captures observed AOD regarding both the mean value as well as the decline rate of the logarithm of AOD over the Atlantic Ocean along the transport path (AOaTP), implying the consistence

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35 with observed export efficiency of African dust along the trans-Atlantic transport.  
36 With an annual emission of  $0.73 \pm 0.12 \text{ Pg } \text{ayr}^{-1}$ , African dust entering the Amazon  
37 Basin has surface concentrations of  $5.7 \pm 1.3 \mu\text{g m}^{-3}$  (up to  $15 \mu\text{g m}^{-3}$  in the northeast  
38 corner) during the wet season, accounting for ~~47% ± 40% ± 4.5.0%~~ (up to 70%) of  
39 mass concentrations of total aerosols. Observed dust peaks over the Amazon Basin are  
40 generally associated with relatively higher African dust emissions (including Sahara  
41 and Sahel) and longer lifetime of dust along the trans-Atlantic transport, namely  
42 higher export efficiency of African dust across the Atlantic Ocean. The frequency of  
43 dust events in the Amazon Basin in the wet season is around 18% averaged over the  
44 basin, with maxima over 60% at the northeast coast. During the dust events, AOD  
45 over most of the Amazon Basin is dominated by dust. ~~Observed peaks of African dust~~  
46 ~~over the Amazon Basin are generally associated with relatively higher African dust~~  
47 ~~emissions and longer lifetime of dust along the trans-Atlantic transport, namely higher~~  
48 ~~export efficiency of African dust across the Atlantic Ocean.~~ Associated with dust  
49 deposition, we further estimate annual inputs of  $52 \pm 8.7$ ,  $0.97 \pm 0.16$  and  $21 \pm 3.6 \text{ mg}$   
50  $\text{m}^{-2} \text{ayr}^{-1}$  for iron, phosphorus and magnesium deposited into the Amazon rainforest,  
51 respectively, which may well compensate the hydrologic losses of nutrients in the  
52 forest ecosystem.

## 54 1 Introduction

55 The desert over North Africa, being the world's largest dust source, contributes to  
56 over 50% of global dust emission (Kok et al., 2021; Wang et al., 2016). Dust particles  
57 are uplifted by strong surface winds, and then typically transported ~~westwards to the~~  
58 ~~tropical downwind for a long distance, reaching~~ Atlantic, Caribbean ~~and~~ America, ~~and~~  
59 Europe (Prospero et al., 1981; Ben-Ami et al., 2012; Yu et al., 2019; Swap et al.,  
60 1992; Prospero et al., 2014); Wang et al., 2020). The emission varies on daily to  
61 seasonal and even decadal time scales, largely affected by local wind speed, land  
62 surface cover, soil moisture, etc (Ridley et al., 2014; Mahowald et al., 2006). Once  
63 present in the atmosphere, mineral dust can degrade air quality downwind as well as

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64 modify the atmospheric radiative balance via directly scattering and absorbing solar  
65 radiation (Ryder et al., 2013b), and altering cloud properties by acting as cloud  
66 condensation nuclei or ice nuclei (Chen et al., 1998; Demott et al., 2003; Mahowald  
67 and Kiehl, 2003; Dusek et al., 2006). Additionally, mineral dust contains iron,  
68 phosphorous, and other nutrients, and could affect ocean biogeochemistry and fertilize  
69 tropical forest upon downwind deposition (Niedermeier et al., 2014; Rizzolo et al.,  
70 2017).

71 There is an increased concern about the impact of African dust exerted over the  
72 Amazon basin, which being the world's largest rainforest, represents a valuable but  
73 also vulnerable ecosystem, and is sensitive to any disturbance from climate changes  
74 associated with human activities in the future (Andreae et al., 2015; Pöhlker et al.,  
75 2019). During the wet season (January – April) Amazonian aerosols are generally  
76 dominated by local biogenic aerosol, with remarkably low PM<sub>10</sub> mass concentrations  
77 of a few  $\mu\text{g m}^{-3}$  (Andreae et al., 2015; Martin et al., 2010a; Prass et al., 2021). The  
78 near-pristine condition, however, is frequently interrupted by the transatlantic  
79 transport of ~~Afreian~~African dust toward the Amazon Basin (Andreae et al., 2015;  
80 Martin et al., 2010b; Martin et al., 2010a; Talbot et al., 1990). The dusty episodes  
81 could drastically increase the aerosol optical depth (AOD, by a factor of 4), mass  
82 concentrations of coarse aerosol (with diameter  $> 1 \mu\text{m}$ ) (up to  $100 \mu\text{g m}^{-3}$ ), as well  
83 as crustal elements over the Amazon Basin (Ben-Ami et al., 2010; Pöhlker et al.,  
84 2019; Moran-Zuloaga et al., 2018; Baars et al., 2011; Formenti et al., 2001).  
85 Therefore, there is great interest in understanding factors controlling the export of  
86 African dust towards the Amazon Basin and the impact they might have on the  
87 environment, ecosystem, and climate.

88 Over the past decades, field measurements combined with satellite observation and  
89 forward/back trajectory analysis have been conducted to explore the long-range  
90 transport (LRT) of African dust toward the Amazon Basin (e.g. Ben-Ami et al., 2010;  
91 Pöhlker et al., 2018; Prospero et al., 2020). The transatlantic transport of African dust  
92 plumes is closely related to annual north-south oscillation of the intertropical

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93 convergence zone (ITCZ) (Moran-Zuloaga et al., 2018; Ben-Ami et al., 2012),  
94 favoring the path towards the Amazon Basin in the late boreal winter and spring  
95 (December-April) as the ITCZ moves southward. In addition to the annual oscillation  
96 of ITCZ, the export efficiency of African dust towards the Amazon Basin also highly  
97 depends on the lifetime of mineral dust, which is largely affected by the  
98 meteorological condition (e.g. precipitation). Dust particles are subject to wet removal  
99 when they are within or underneath precipitating clouds. For instance, Yu et al. (2020)  
100 argued that El Djouf, in western Sahara, contributes more dust to the Amazon Basin  
101 than the Bodélé depression as the transport paths of dust released from El Djouf are  
102 less affected by the rainy cloud.

103 Besides meteorological conditions, dust size distribution can also influence its  
104 lifetime and consequently the export efficiency of African dust towards the Amazon  
105 Basin. Previous studies have observed that volume/mass fractions of coarse mode dust  
106 particles, giant particles in particular, tend to be reduced along the transport due to  
107 higher gravitational settling velocities (Ryder et al., 2018; Ryder et al., 2013b; Ryder  
108 et al., 2013a; Van Der Does et al., 2016). Moreover, the optical properties of mineral  
109 dust are also strongly size dependent, especially for those in sub-micron range (Liu et  
110 al., 2018; Di Biagio et al., 2019; Ysard et al., 2018). For instance, Ryder et al. (2013a)  
111 reported a loss of 60 – 90% of particles with diameter > 30  $\mu\text{m}$  ~~in size~~ 12 h after uplift  
112 and consequently an increase in the single scattering albedo from 0.92 to 0.95  
113 associated with the change in the size distribution of dust aerosols. Therefore, the size  
114 distribution of dust particles is a key factor determining the efficiency of dust  
115 transport and consequently the environmental and climate effect of the mineral dust  
116 downwind (Mahowald et al., 2011a; Mahowald et al., 2011b).

117 It is worth ~~mention~~mentioning that the LRT events bring not only mineral dust into  
118 the Amazon Basin but also biomass burning aerosols from Africa as well as sea spray  
119 aerosols (Wang et al., 2016; Holanda et al., 2020; Andreae et al., 1990; Talbot et al.,  
120 1990; Ansmann et al., 2009; Baars et al., 2011), making it challenging to have a  
121 quantitative assessment of the impact of African dust on the Amazon Basin. So far, a

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122 few studies have attempted to quantify the impact of the LRT of African dust over the  
123 Amazon Basin, but mainly focus on dust deposition only (e.g. Yu et al., 2015a; Ridley  
124 et al., 2012; Yu et al., 2019). Estimates of annual dust deposition and dust deposition  
125 rates into the Amazon Basin exhibit a wide range (7.7-50 Tg ~~ayr~~<sup>-1</sup> and 0.8-19 g m<sup>-2</sup>  
126 ~~ayr~~<sup>-1</sup>, respectively), attributed to the application of different methods and the intrinsic  
127 uncertainties associated with each method (Kok et al., 2021; Yu et al., 2015b;  
128 Kaufman, 2005; Swap et al., 1992). For example, the results based on Cloud-Aerosol  
129 Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) is subject to the  
130 uncertainty associated with the Cloud-Aerosol Lidar with Orthogonal Polarization  
131 (CALIOP) extinction, vertical profile shape, dust discrimination, diurnal variations of  
132 dust transport as well as the below-cloud dust missed by CALIOP (Yu et al., 2015a).  
133 While models could be considered as a useful tool to comprehensively assess the  
134 transatlantic transport of African dust towards the Amazon Basin and the consequent  
135 impact over the Amazon Basin, there exist considerable differences among model  
136 results, attributed to the uncertainties associated with the dust parameterization in the  
137 model, including emission schemes, dust size distribution, dust deposition, etc (Kim  
138 et al., 2014; Huneeus et al., 2011; Mahowald et al., 2014). Observational constraints  
139 on the modelling results along the transport from source regions to receptor regions  
140 are thus ~~necessarily needed to demonstrate the model performance and~~ in urgent need  
141 to accomplish a better evaluation of factors controlling the ~~variability in the~~ LRT of  
142 African dust and its overall impact over the Amazon Basin ~~assessment~~.  
143 Here, we present a detailed multiyear simulation of the export of ~~Africa~~ African dust  
144 across the Atlantic and ~~theits~~ impact over the Amazon Basin (around  $8.8 \times 10^6$  km<sup>2</sup>,  
145 see ~~Fig.~~ Figure 1 for defined area) during 2013 – 2017 with the GEOS-Chem  
146 (chemical transport model). The aims of this study are: (1) to evaluate the model  
147 performance ~~regarding theof dust~~ simulation-of-dust-aerosols including the particle  
148 mass size distribution (PMSD), optical properties, mass concentrations as well as the  
149 ~~trans-Atlantic-transport~~ export efficiency of African dust towards the Amazon Basin;  
150 (2) to analyze factors controlling the export of African dust towards the Amazon

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151 Basin; and (3) to give a comprehensive examination of the impact of African dust  
152 over the Amazon Basin, including surface aerosol concentrations, AOD and nutrient  
153 inputs upon deposition.

## 154

## 155 2 **2-Methodology**

### 156 **2.1- GEOS-Chem model**

#### 157 **2.1.1 Model overview**

158 In this study we use the GEOS-Chem model version 12.0.0 (www. geos-chem.org) to  
159 perform a global aerosol simulation with a horizontal resolution of  $2^\circ \times 2.5^\circ$ . The  
160 GEOS-Chem is driven by assimilated meteorological data GEOS-FP from the NASA  
161 Global Modeling and Assimilation Office (GMAO) (Lucchesi, 2013) with a native  
162 horizontal resolution of  $0.25^\circ \times 0.3125^\circ$ , which is then degraded to  $2^\circ \times 2.5^\circ$  for the  
163 input to GEOS-Chem. We initialize the model with a 1-year spin-up followed by an  
164 aerosol simulation from 2013 to 2017.

165 The aerosol simulation is an offline simulation for aerosol tracers including ~~mineral~~  
166 ~~dust in four size bins, sea salt in fine ( $\leq 1 \mu\text{m}$  in diameter) and coarse ( $> 1 \mu\text{m}$  in~~  
167 ~~diameter) modes,~~ black carbon (BC), organic aerosols (OA), and sulfate-nitrate-  
168 ammonium aerosols in fine mode ( $\leq 1 \mu\text{m}$  in diameter), sea salt in both fine and  
169 coarse ( $> 1 \mu\text{m}$  in diameter) modes, and mineral dust in four size bins covering the  
170 size range of  $0.2 - 12 \mu\text{m}$  in diameter. Aerosol optical properties used for aerosol  
171 optical depth (AOD) calculation are mainly based on Global Aerosol Data Set  
172 (Koepke et al., 1997), with modifications in aerosol size distributions (Jaeglé et al.,  
173 2011; Drury et al., 2010; Wang et al., 2003a; Wang et al., 2003b), hygroscopic growth  
174 factor of organic aerosols (Jimenez et al., 2009), and the refractive index of dust  
175 (Sinyuk et al., 2003). AOD in the model is then calculated online at selected  
176 wavelengths assuming lognormal size distributions of externally mixed aerosols and  
177 accounts for hygroscopic growth (Martin et al., 2003).

178 Wet deposition in GEOS-Chem, based on the scheme of Liu et al. (2001), accounts for  
179 scavenging in both convective updrafts and large-scale precipitation. Further updates

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180 by Wang et al. (2011) are also applied, accounting for ice/snow scavenging as well as  
181 the impaction scavenging in convective updrafts. Dry deposition in the model follows  
182 the standard resistance-in-series scheme by Wesely (2007), accounting for turbulent  
183 transfer and gravitational settling (Wang et al., 1998; Zhang et al., 2001).

### 184 2.1.2 Dust emission and PMSD schemes in the model

#### 185 ~~2.2 Observations~~

186 ~~The particle volume size distribution (PVSD) and AOD daily data (at wavelength of~~  
187 ~~675 nm) from AERONET level 2.0~~  
188 ~~([aeronet.gsfc.nasa.gov/new\\_web/download\\_all\\_v3\\_aod.html](http://aeronet.gsfc.nasa.gov/new_web/download_all_v3_aod.html), last access on Jun. 22,~~  
189 ~~2021 (Dubovik et al., 2002)) during the year of 2013–2017 is used in the study to~~  
190 ~~evaluate dust emissions and its PMSD over the source regions in Africa. The PVSD~~  
191 ~~data provided by AERONET is a column-integrated aerosol volume size distribution~~  
192 ~~and with a size range of 0.05–15.0  $\mu\text{m}$ . For comparison with model results, the~~  
193 ~~PVSD data is converted to PMSD using the same densities as in the model. In~~  
194 ~~addition, to minimize the influence of aerosols other than dust, only data dominated~~  
195 ~~by coarse aerosol (contribution of fine aerosol to total aerosol volume  $< 3\%$  or~~  
196 ~~simulated dust contribution  $> 95\%$ ) is used for the comparison.~~

197 ~~The study also uses observed PMSD over central Sahara during Fennec Campaign~~  
198 ~~([africanclimateoxford.net/projects/fennec/](http://africanclimateoxford.net/projects/fennec/), last access: 22 June 2021) for the~~  
199 ~~comparison with AERONET and our model results. Aiming to investigate dust~~  
200 ~~microphysical and optical properties, 42 profiles of size distribution (0.1–300  $\mu\text{m}$  in~~  
201 ~~diameter) over both the Sahara and the Atlantic Ocean, were sampled from in-situ~~  
202 ~~aircraft measurements during Fennec campaign. For more detailed description of the~~  
203 ~~aircraft measurements, readers are referred to Ryder et al. (2013a).~~

204 ~~In addition, level 3 daily AOD (at wavelength of 550 nm) data from the moderate~~  
205 ~~resolution imaging spectroradiometers (MODIS) installed on Terra and Aqua~~  
206 ~~platforms (<https://ladsweb.modaps.eosdis.nasa.gov/archive/allData/61/>, last access: 22~~  
207 ~~June 2021) is applied in the study to evaluate the trans-Atlantic transport of dust~~  
208 ~~plumes from Africa toward Amazon Basin. For comparison, we degraded the original~~



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209 ~~horizontal resolution of MODIS data ( $1^\circ \times 1^\circ$ ) to  $2^\circ \times 2.5^\circ$ , consistent with the model~~  
210 ~~outputs.~~

211 ~~Finally, long-term aerosol measurements at the Amazon Tall Tower Observatory~~  
212 ~~(ATTO,  $59.0056^\circ$  W,  $2.1459^\circ$  S, marked in Fig. 1), located in central Amazon Basin~~  
213 ~~are used in the study to evaluate the influence of the long-range transport of African~~  
214 ~~dust over the Amazon Basin. The ATTO site was established in 2012 for the long-term~~  
215 ~~monitoring of climatic, biogeochemical, and atmospheric conditions in the Amazon~~  
216 ~~rainforest. Detailed description of the site and the measurements there could be found~~  
217 ~~in Andreae et al. (2015). In this study, we only use the particle number size~~  
218 ~~distribution from a Optical Particle Sizer (OPS, TSI model 3330; size range of 0.3~~  
219  ~~$\mu\text{m}$  in diameter, divided into 16 size bins) and a Scanning Mobility Particle Sizer~~  
220 ~~(SMPS, TSI model 3080, St. Paul, MN, USA; size range of 10–430 nm in diameter,~~  
221 ~~divided into 104 size bins) over the period from 2014 to 2016. The number size~~  
222 ~~distribution is converted to mass concentrations assuming spherical particles with a~~  
223 ~~constant density of  $1.5 \text{ g cm}^{-3}$  (Pöschl et al., 2010). In addition, daily  $\text{PM}_{10}$  mass~~  
224 ~~concentrations during wet season (from January to April) in 2014 measured at~~  
225 ~~Cayenne, French Guiana ( $4.9489^\circ$  N,  $52.3097^\circ$  W, located in the northeast coast of the~~  
226 ~~Amazon Basin, marked in Fig. 1, <https://doi.org/10.17604/vrsh-w974>) are also used in~~  
227 ~~this study to further evaluate the model performance regarding the trans-Atlantic~~  
228 ~~transport of African dust toward the Amazon basin. The measurement at Cayenne site~~  
229 ~~is carried out on a cooperative basis by personnel of ATMO-Guyane, a non-profit~~  
230 ~~organization (<https://www.atmo-guyane.org/qui-sommes-nous/statuts/>). The  $\text{PM}_{10}$~~   
231 ~~samples are measured by a Taper Element Oscillating Microbalance (TEOM, model~~  
232 ~~1400 series, ThermoFisher Scientific) and then are returned to Miami for analysis~~  
233 ~~(Prospero et al., 2020). Readers are referred to Prospero et al. (2020) for detailed~~  
234 ~~description of the site and the data.~~

### 235

## 236 **3 Dust emissions and size distribution**

### 237 **3.1 Dust emissions**



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238 The emission of mineral dust is based on the dust entrainment and deposition (DEAD)  
239 mobilization scheme of Zender et al. (2003) in the GEOS-Chem model (Duncan  
240 Fairlie et al., 2007). Figure 1 shows the spatial distribution of simulated dust  
241 emissions and Table 1 lists seasonal and annual dust emissions in northern Africa  
242 ( $17.5^{\circ} \text{W} - 40^{\circ} \text{E}$ ,  $10^{\circ} \text{N} - 35^{\circ} \text{N}$ ) for the period of 2013 – 2017. Simulated annual  
243 dust emission from northern Africa is  $0.73 \pm 0.12 \text{ Pg a}^{-1}$ , contributing more than 70%  
244 of the global dust emission ( $0.99 \pm 0.20 \text{ Pg a}^{-1}$ ). The result is in the range of  $0.42 -$   
245  $2.05 \text{ Pg a}^{-1}$  reported by Kim et al.. The DEAD scheme calculates the total vertical  
246 dust flux based on the total horizontal saltation flux ( $Q_s$ ) using the theory of White  
247 (1979). The  $Q_s$  depends mainly on the surface wind friction velocity and the threshold  
248 friction velocity, which is determined by soil type, soil moisture content, and surface  
249 roughness. For more details of the DEAD scheme, readers are referred to Duncan  
250 Fairlie et al. (2007), (2014), who evaluated five AeroCom II global models regarding  
251 the dust simulation over similar regions.

252 There exists a strong seasonality in the dust emission from northern Africa, peaking in  
253 spring and winter (up to  $1.2 \text{ Pg a}^{-1}$ ) and reaching the minimum in fall (around  $0.33 \text{ Pg}$   
254  $\text{a}^{-1}$ ) in general. Previous studies have also pointed out that dust emissions over  
255 different African regions show distinct variations (Bakker et al., 2019; Shao et al.,  
256 2010), attributed to differences in geographical properties (Knippertz et al., 2007),  
257 vegetation cover (Mahowald et al., 2006; Kim et al., 2017), and meteorological  
258 mechanisms on a local scale (Alizadeh Choobari et al., 2014; Wang et al., 2017;  
259 Roberts and Knippertz, 2012). Consequently, there exists substantial seasonal change  
260 in dust source regions. For instance, during boreal winter, the Bodélé Depression in  
261 northern Chad is found to be the most active triggered by the Harmattan winds, while  
262 the northwestern African dust sources become less active in contrast with the  
263 condition in boreal summer (Ben-Ami et al., 2012; Prospero et al., 2014). Therefore,  
264 we further analyze the emission variability over five different source regions in  
265 northern Africa (Fig. 1 and Table S1). On an annual basis, the contribution to total  
266 northern African dust emission is the largest from Region A (west Sahara,  $36\% \pm$

267 4.0%), followed by Region D (central Sahel including Bodélé,  $21\% \pm 4.7\%$ ), Region  
268 B (central Sahara,  $13\% \pm 2.6\%$ ), Region C (east Sahara,  $12\% \pm 1.0\%$ ), and Region E  
269 (west Sahel,  $6.5\% \pm 0.64\%$ ). The emission fluxes, however, are the most intensive in  
270 Region D, up to  $11 \pm 2.1 \text{ g month}^{-1} \text{ m}^{-2}$  and are generally below  $5 \text{ g month}^{-1} \text{ m}^{-2}$  over  
271 the other regions. Concerning the seasonality, higher dust emission tends to occur in  
272 boreal spring and winter, with the largest emission flux of  $19 \pm 4.7 \text{ g month}^{-1} \text{ m}^{-2}$  from  
273 Region D. As shown in Fig. 2 and S1, the emissions peak in boreal spring for Region  
274 A, B and C, but in winter for Region D and E. There is also a secondary peak in  
275 summer emissions for Region E. Correlation analysis between dust emissions and  
276 meteorological variables suggests that the seasonality is mainly driven by high surface  
277 wind speeds (with  $r$  of 0.79–0.96 and 0.68–0.97 for the 75<sup>th</sup> and 95<sup>th</sup> percentiles of  
278 wind speeds, respectively). Apparent negative correlation is also found between  
279 precipitation (soil moisture, Fig. S2) and dust emission in Region D with  $r$  of  $-0.69$  ( $-$   
280  $0.67$ ). Similar seasonality is also reported by Cowie et al. (2014), who suggested that  
281 the strongest dust season in winter in central Sahel is driven by strong harmattan  
282 winds and frequent Low level Jet breakdown, and the second peak in summer in west  
283 Sahel could be explained by the summer monsoon combined with the Sahara Heat  
284 Low. The study also suggested the dominance of strong wind frequency in the  
285 seasonal variation of the emission frequencies.  
286 There is a significant decrease in the annual emission from  $0.88 \text{ Pg a}^{-1}$  in 2013 to  $0.56$   
287  $\text{Pg a}^{-1}$  in 2017. Similarly, studies on African dust variability at decadal and longer time  
288 scales also reported an obviously decreasing trend in both dust emissions in Africa  
289 and dustiness over the east mid-Atlantic in recent decades since the early 1980s  
290 (Ridley et al., 2014; Middleton, 2019; Shao et al., 2013). Evan et al. (2016) pointed  
291 out three periods of persistently low dust concentrations in the 1860s,  
292 1950s and 2000s. Weather and climate drivers behind this variability include  
293 precipitation, surface wind over northern Africa, Atlantic Multidecadal Oscillation  
294 (AMO), North Atlantic Oscillation (NAO), the movement of ITCZ, etc. For example,  
295 as shown in Fig. S2, there is an obviously increasing trend of AMO over the period

296 2000–2015, especially from 2010 to 2015 (data available from [http://www.esrl-  
298 noaa.gov](http://www.esrl-<br/>297 noaa.gov), last access on July 29, 2021 (Enfield et al., 2001)). This positive AMO  
299 phase corresponds to higher North Atlantic sea surface temperature (SST), and could  
300 result in enhanced rainfall in the Sahel and consequently less African dust emissions  
301 (Middleton, 2019). A recent study by Yuan et al. (2020) projected decreased surface  
302 wind speeds over African dust source regions as well as more precipitation in the  
303 Sahel region due to positive interhemispheric contrast in Atlantic SST associated with  
304 the global warming, leading to less dust emissions and weaker westward transport.  
305 While most regions show decline trends of dust emissions, the emission in Region D  
306 shows a slight increase. The variation is mainly associated with surface wind speeds  
307 (Fig. S3). For instance, the 75<sup>th</sup> and 95<sup>th</sup> percentiles of wind speeds decrease by 7.0%  
308 and 9.1% in Region B but slightly increases by 2.0% and 1.4% in Region D from  
309 2013 to 2017. The  $r$  values are in the range of 0.90–0.99 between annual dust  
310 emissions and the 95<sup>th</sup> percentile of wind speeds over all the 5 source regions.  
311 Significant negative correlation with  $r$  of  $-0.52$ – $-0.73$  between annual dust emissions  
312 and soil moisture is also found for those regions except for Region D where  $r$  is only  
313 0.08.

314 It is also worth noting that the interannual variation in dust emission is much larger  
315 during the wet season ( $0.96 \pm 0.25$ , Table 1) than on an annual basis ( $0.73 \pm 0.12$ ).  
316 Moreover, while the annual emissions gradually decrease from 2013 to 2017, the  
317 emissions during the wet season peak in 2015. The obviously different behavior  
318 between the annual emissions and emissions during the wet season suggests that  
319 predictions of future impact of African dust emissions over the Amazon Basin in  
320 response to climate change should focus on the wet season rather than the annual  
321 average, as the former is more related to the export of African dust towards the  
322 Amazon Basin.

### 323 **3.2 Dust size distribution and its impact on the export efficiency towards the** 324 **Amazon Basin**

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325 Freshly emitted dust particles are divided into four size bins in GEOS-Chem: 0.1 –  
326 1.0  $\mu\text{m}$ , 1.0 – 1.8  $\mu\text{m}$ , 1.8 – 3.0  $\mu\text{m}$ , and 3.0 – 6.0  $\mu\text{m}$  in radius. The first size bin is  
327 further divided into four sub-bins (0.1 – 0.18  $\mu\text{m}$ , 0.18 – 0.3  $\mu\text{m}$ , 0.3 – 0.6  $\mu\text{m}$ , and  
328 0.6 – 1.0  $\mu\text{m}$  in radius) for the calculation of optical properties. While total dust  
329 emissions are not affected, optical properties, atmospheric lifetime and downwind  
330 concentrations of dust particles are sensitive to different PMSD schemes.

331 Table 21 shows 3 different PMSD schemes tested in this study: V12, V12\_C and  
332 V12\_F. Scheme V12, which is derived based on scale-invariant fragmentation theory  
333 (Kok, 2011) with modification in tunable parameters (Zhang et al., 2013), is a default  
334 set in GEOS-Chem. However, this scheme has been only evaluated for US/Asian dust,  
335 not for Africa. On the other hand, V12\_C was used in older versions of GEOS-Chem  
336 and constrained from aircraft measurements during the Saharan Dust Experiment  
337 (Ridley et al., 2012; Highwood et al., 2003). In addition, we derived V12\_F based on  
338 the measurements during the Fennec aircraft observations also focusing on Saharan  
339 dust. Among all the three PMSD, V12\_C has the largest mass fraction in the first bin  
340 (relatively small particles) and the lowest fraction in the last bin (large ones). In  
341 contrast, V12\_F has the most dust distributed in the last bin ( $\sim 70\%$ ) and only a little  
342 (around 5%) in the first bin (0.1 – 1.0  $\mu\text{m}$ ).

343 Simulated mass extinction efficiency (MEE, also shown in Table 21) at wavelength of  
344 550 nm for dust particles in the first sub-bin (0.1 – 0.18  $\mu\text{m}$ ) is  $3.13 \text{ m}^2 \text{ g}^{-1}$ ; and  
345 decreases to  $0.16 \text{ m}^2 \text{ g}^{-1}$  for those in the last bin (3.0 – 6.0  $\mu\text{m}$ ). The lifetime of dust  
346 aerosols against deposition are 5.1, 2.2, 1.7 and 0.86 d in the four bins (from small to  
347 large size), respectively. Therefore, ~~as mentioned before,~~ although with the same  
348 emission, total dust AOD, lifetime, and downwind concentrations could vary greatly  
349 with PMSD. ~~Here upon emissions. In this study, we will evaluate all-these~~ three  
350 PMSD schemes and the impact on AOD, dust concentrations as well as its export  
351 efficiency along the trans-Atlantic transport from Africa to the Amazon Basin.

## 352 2.2 Observations

353 The study uses observations from multiple datasets obtained from AERONET,

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354 MODIS, as well as Cayenne site and the Amazon Tall Tower Observatory (ATTO) site  
355 to constrain model results regarding the simulation of the dust export from Africa to  
356 the Amazon Basin. Table 2 summarizes these observations, including the parameters,  
357 the spatio-temporal coverage, and the corresponding application in the model. The  
358 daily data of AOD (at wavelength of 675 nm) and particle volume size distribution  
359 (PVSD) from AERONET level 2.0  
360 ([aeronet.gsfc.nasa.gov/new\\_web/download\\_all\\_v3\\_aod.html](http://aeronet.gsfc.nasa.gov/new_web/download_all_v3_aod.html), last access on Jun. 22,  
361 2021(Dubovik et al., 2002)) during the year of 2013 – 2017 is used in the study to  
362 evaluate dust emissions and its PMSD over the source regions in Africa in the model.  
363 The PVSD data provided by AERONET is a column-integrated aerosol volume size  
364 distribution and with a size range of 0.05 – 15.0  $\mu\text{m}$ . It is then converted to PMSD  
365 using the same densities as in the model. Only sites with valid data accounting for  
366 more than 30% of the total are considered in this study. In addition, to minimize the  
367 influence of aerosols other than dust, only data dominated by dust (simulated dust  
368 contribution to column-integrated aerosols mass concentrations > 95%) is used for the  
369 comparison of PMSD. There are a few sites not far from the coast and could be  
370 influenced by sea salt. With the above data screening, the sea salt contribution to total  
371 aerosol mass is less than 0.5%. For the comparison of AOD, the criterion is less  
372 stringent to have more data points available and uses data dominated by coarse  
373 aerosols (the contribution of fine aerosol to total aerosol volume < 3%). This criterion  
374 does not exclude sea salt and the contribution of sea salt to AOD could be up to 30%  
375 at the Capo Verde site (22.9° W, 16.7° N) over the east of the Atlantic Ocean.  
376 The study also uses observed PMSD over central Sahara during Fennec Campaign  
377 ([africanclimateoxford.net/projects/fennec/](http://africanclimateoxford.net/projects/fennec/), last access: 22 June 2021) for the  
378 comparison with AERONET and our model results. Aiming to investigate dust  
379 microphysical and optical properties, 42 profiles of size distribution (0.1 – 300  $\mu\text{m}$  in  
380 diameter) over both the Sahara and the Atlantic Ocean, were sampled from in-situ  
381 aircraft measurements during Fennec campaign. For more detailed description of the  
382 aircraft measurements, readers are referred to Ryder et al. (2013a).

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383 In addition to AERONET AOD data, level 3 daily AOD (at wavelength of 550 nm)  
384 data from the moderate resolution imaging spectroradiometers (MODIS) installed on  
385 Terra and Aqua platforms  
386 (<https://ladsweb.modaps.eosdis.nasa.gov/archive/allData/61/>, last access: 22 June  
387 2021) is applied in the study to evaluate the trans-Atlantic transport of dust plumes  
388 from Africa toward Amazon Basin. For comparison, we degraded the original  
389 horizontal resolution of MODIS data ( $1^\circ \times 1^\circ$ ) to  $2^\circ \times 2.5^\circ$ , consistent with the model  
390 outputs.

391 Finally, daily PM<sub>10</sub> mass concentrations during wet season (from January to April) in  
392 2014 measured at Cayenne, French Guiana ( $52.3097^\circ$  W,  $4.9489^\circ$  N, located in the  
393 northeast coast of the Amazon Basin, <https://doi.org/10.17604/vrsh-w974>, marked in  
394 Figure 1) and long-term aerosol measurements at the Amazon Tall Tower  
395 Observatory, Brazil (ATTO,  $59.0056^\circ$  W,  $2.1459^\circ$  S, located in central Amazon Basin,  
396 also marked in Figure 1) are used in this study to further evaluate the model  
397 performance regarding the influence of the LRT of African dust over the Amazon  
398 Basin. The measurement at Cayenne site is carried out on a cooperative basis by  
399 personnel of ATMO-Guyane, a non-profit organization. The PM<sub>10</sub> samples are  
400 measured by a Taper Element Oscillating Microbalance (TEOM, model 1400 series,  
401 ThermoFisher Scientific) and then are returned to Miami for analysis (Prospero et al.,  
402 2020). Readers are referred to Prospero et al. (2020) for detailed description of the site  
403 and the data. The ATTO site was established in 2012 for the long-term monitoring of  
404 climatic, biogeochemical, and atmospheric conditions in the Amazon rainforest.  
405 Detailed description of the site and the measurements there could be found in Andreae  
406 et al. (2015). In this study, we only use the particle number size distribution from an  
407 Optical Particle Sizer (OPS, TSI model 3330; size range of 0.3 – 10  $\mu$ m in diameter,  
408 divided into 16 size bins) and a Scanning Mobility Particle Sizer (SMPS, TSI model  
409 3080, St. Paul, MU, USA; size range of 10 – 430 nm in diameter, divided into 104  
410 size bins) over the period from 2014 to 2016. The number size distribution is  
411 converted to mass concentrations assuming spherical particles with a constant density

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412 of  $1.5 \text{ g cm}^{-3}$  (Pöschl et al., 2010).

### 414 3 Model evaluation

415 Here we evaluate three different PMSD schemes regarding the model performance of  
416 dust simulation through the comparison with observed mass size distribution of  
417 column-integrated aerosol over Africa, AOD over both Africa and the Atlantic Ocean,  
418 as well as PM<sub>10</sub> and dust concentrations in the Amazon Basin.

419 Figure 32 shows the mass fractions of column-integrated aerosols retrieved from  
420 AERONET sites compared with model results based on different PMSD schemes. The  
421 location of the selected AERONET sites with valid data are marked in Fig-Figure 1 as  
422 purple symbols (including asterisks and circles). The mean mass fractions for each bin  
423 from AERONET data are 17%, 27%, 38%, 17%, respectively. The comparison  
424 indicates the model results based on V12\_C agrees better with the observations, with  
425 much smaller mean absolute deviation (MAD) of 2.8, followed by 4.2 for V12 and 18  
426 for V12\_F. In other words, the model results with other PMSD schemes (V12\_F in  
427 particular) greatly underestimate the mass fractions in the first bin and overestimate  
428 the mass fractions in the last bin. During the Fennec campaign, the aircraft sampled  
429 two strong Saharan dust outbreaks with AOD up to 1.1, which may be associated with  
430 strong winds favoring the uplift of large particles.

431 Figure 43 shows the times series of daily AOD at wavelength of 675 nm during the  
432 year of 2013 – 2017 from both AERONET and model results. The locations of the  
433 selected AERONET sites with valid data over northern Africa are shown in Fig-Figure  
434 1 as purple circles. The Capo Verde site over the east of the Atlantic Ocean is also  
435 included to show the model performance over the ocean in addition to the land.

436 Although different PSD schemes have little influence on the correlation between  
437 AERONET and model results with most  $r$  around 0.6 – 0.7, the normalized mean bias  
438 (NMB) has been significantly improved in V12\_C, with a range of -12% – 11% (vs. -  
439 33% – -11% and -42% – -19% for V12 and V12\_F, respectively). The severe  
440 underestimation in AOD from V12 and V12\_F could be attributed to their relatively



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441 higher dust fractions distributed in larger size bins with lower MEE.

442 In addition, we also compare the spatial distributions of simulated AOD over the

443 Atlantic Ocean with MODIS AOD (at 550 nm) averaged over 2013 – 2017 in ~~Fig.~~

444 ~~5a~~Figure 4a-d. There is a clear decreasing trend in MODIS AOD along the trans-

445 Atlantic transport from Africa towards South America. Although all simulations show

446 similar spatial distributions with declining trends of AOD along the transport, the

447 results from V12\_C are the most consistent with MODIS data with the highest  $r$  of

448 0.89 and the smallest NMB of 6.5% among the three schemes (vs.  $r$  of 0.85 and 0.81

449 and NMB of -13% and -19% for V12 and V12\_F, respectively). Note that the model

450 results based on V12\_C tends to overestimate MODIS AOD over Africa while no

451 significant systematic bias is found between V12\_C and AERONET AOD. Wang et

452 al. (2016) sampled MODIS data at AERONET sites over Africa and found that

453 MODIS retrieval underestimate AERONET AOD at most sites with NMB of -12% –

454 36%, which partly explain the large difference between model V12\_C and MODIS

455 AOD.

456 Assuming first-order removal of aerosol along the transport, we ~~derived linear trend~~

457 ~~lines based on~~could derive the removal rates of aerosols, estimated as the gradient of

458 the logarithm of AOD ( $\log(\text{AOD})$ ) against the distance over the Atlantic Ocean along

459 the transport path (AOaTP, 20° – 50° W and 5° S – 25° N, ~~Fig. 5e~~). ~~MODIS AOD~~

460 ~~decrease from  $0.29 \pm 0.023$  near the coast of Africa to  $0.17 \pm 0.010$  at the coast of~~

461 ~~South America, with a~~Figure 4e). The decline rate of MODIS  $\log(\text{AOD})$  is  $0.019 \pm$

462  $0.0025 \text{ degree}^{-1}$ . A similar decline rate of  $0.019 \pm 0.0029 \text{ degree}^{-1}$  is found for

463 simulated  $\log(\text{AOD})$  based on V12\_C, decreasing from  $0.28 \pm 0.022$  to  $0.16 \pm 0.013$

464 ( $0.019 \pm 0.0029 \text{ degree}^{-1}$ ). In contrast, simulations with V12 and V12\_F exhibit ~~much~~

465 ~~lower AOD together with~~ relatively steeper slopes of  $0.021 \pm 0.0040$  and  $0.021 \pm$

466  $0.0041$ , respectively, implying too much aerosol removal and thus lower export

467 efficiency along the transport. To specify the impact of different PMSD on the export

468 efficiency of dust aerosols towards the Amazon Basin, ~~Fig. 5f~~Figure 4f also shows

469 simulated dust AOD (DOD) along the transect from 20° to 50° W. The DOD from

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470 V12\_C decreases from  $0.15 \pm 0.018$  to  $0.049 \pm 0.088$  along the transport, with a  
471 decreasing rate of  $0.016 \pm 0.0014$  degree<sup>-1</sup>. In contrast, DOD decreases from  $0.097 \pm$   
472  $0.012$  to  $0.028 \pm 0.085$  with a slope of  $0.018 \pm 0.0016$  for V12 and decreases from  
473  $0.080 \pm 0.090$  to  $0.025 \pm 0.084$  with a slope of  $0.017 \pm 0.0014$  for V12\_F.  
474 Lying in the trade wind belt, Cayenne has been taken as the gate of African dust.  
475 Hence, the comparison between simulated and observed dust concentrations at  
476 Cayenne site could evaluate model performance in reproducing the arrival of African  
477 dust to the Amazon Basin. As shown in ~~Fig. 6a~~Figure 5a, the simulation from V12\_C  
478 shows excellent agreement between simulated dust and observed PM<sub>10</sub> concentration  
479 during wet season, with  $r$  around 0.85 and NMB of -39%. The correlation from the  
480 other two simulations is similar ( $r = 0.86$ ), but the corresponding NMB is much larger  
481 (-57% for V12 and -80% for V12\_F). ~~Prospero et al. (2020) did similar analysis at the~~  
482 ~~Cayenne site but concerning the data all year round.~~ Based on the regression line  
483 between observed concentrations of PM<sub>10</sub> and dust, ~~they at the same site, Prospero et~~  
484 ~~al. (2020) obtained a regional background value of PM<sub>10</sub> ranging from 17 to 22  $\mu\text{g m}^{-3}$ ,~~  
485 ~~largely attributed to sea salt aerosol, and suggested a value of 0.9 for the slope,~~  
486 ~~suggesting PM<sub>10</sub> values above this range as a proxy for advected dust. Consistent with~~  
487 ~~their results, the regression line between observed PM<sub>10</sub> and simulated dust and PM<sub>10</sub>~~  
488 ~~from V12\_C in this study shows a background value of PM<sub>10</sub> around 23  $\mu\text{g m}^{-3}$ . The~~  
489 ~~slope of the regression line from V12\_C is 1.0, also consistent, with the value of 0.9~~  
490 ~~in the study of Prospero et al. (2020), demonstrating the well performance of the~~  
491 ~~model with V12\_C in simulating the trans-Atlantic transport of African the slope~~  
492 ~~around 1.0, and the dust towards the Amazon Basin. contribution to PM<sub>10</sub> is around~~  
493 ~~53%  $\pm$  20%. In contrast, the regression lines from V12 and V12\_F are much steeper,~~  
494 ~~with the slope of 1.4 and 2.1, respectively, and the dust contributions are~~  
495 ~~relatively smaller, 44% in V12 and 34% in V12\_F.~~  
496 We also compare simulated dust concentrations with observed coarse particles at  
497 ATTO site in central Amazon in wet season during 2014 – 2016 in ~~Fig. 6b~~Figure 5b.  
498 The correlation between observed and simulated data are similar for different PMSD

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499 schemes with  $r$  of 0.63 – 0.65. But the bias of V12\_C is negligible (NMB = -0.27%)  
500 while both V12 and V12\_F tend to underestimate the coarse aerosol concentrations  
501 with NMB of -36% and -55%, respectively. The dust contribution to coarse aerosols  
502 in above 80% in V12\_C, but less than 70% in V12\_F. This again implies relatively  
503 higher export efficiency of African dust aerosols towards the Amazon Basin with  
504 V12\_C associated with their relatively higher dust fractions distributed in smaller size  
505 bins.

506 Overall, compared with V12 and V12\_F schemes, model results based on V12\_C are  
507 more consistent with the multiple observed data sets, including column-integrated  
508 particle size distribution, AOD and surface coarse aerosol concentrations obtained  
509 either over sources or downwind of the sources. Therefore, we use the model results  
510 from V12\_C (hereinafter referred to as model results unless noted otherwise) to  
511 investigate the transatlantic transport of dust from Africa and its impact over the  
512 Amazon Basin in the following sections.

513

## 514 **4 Results and discussion**

### 515 **4.1 Dust emissions**

516 Figure 1 shows the spatial distribution of simulated dust emissions and Table 3 lists  
517 seasonal and annual dust emissions in northern Africa (17.5° W – 40° E, 10° N – 35°  
518 N) for the period of 2013 – 2017. Simulated annual dust emission from northern  
519 Africa is  $0.73 \pm 0.12$  Pg yr<sup>-1</sup>, contributing more than 70% of the global dust emission  
520 ( $0.99 \pm 0.20$  Pg yr<sup>-1</sup>). The result is in the range of 0.42 – 2.05 Pg yr<sup>-1</sup> reported by Kim  
521 et al. (2014), who evaluated five AeroCom II global models regarding the dust  
522 simulation over similar regions.

523 There exists a strong seasonality in the dust emission from northern Africa, peaking in  
524 spring and winter (up to 1.2 Pg yr<sup>-1</sup>) and reaching the minimum in fall (around 0.33  
525 Pg yr<sup>-1</sup>) in general. Previous studies have also pointed out that dust emissions over  
526 different African regions show distinct variations (Bakker et al., 2019; Shao et al.,  
527 2010), attributed to differences in geographical properties (Knippertz et al., 2007),

528 vegetation cover (Mahowald et al., 2006; Kim et al., 2017), and meteorological  
529 mechanisms on a local scale (Alizadeh-Choobari et al., 2014; Wang et al., 2017;  
530 Roberts and Knippertz, 2012). Consequently, there exists substantial seasonal change  
531 in dust source regions. For instance, during boreal winter, the Bodélé Depression in  
532 northern Chad is found to be the most active triggered by the Harmattan winds, while  
533 the northwestern African dust sources become less active in contrast with the  
534 condition in boreal summer (Ben-Ami et al., 2012; Prospero et al., 2014). Therefore,  
535 we further analyze the emission variability over five different source regions in  
536 northern Africa (Figure 1 and Table S1). On an annual basis, the contribution to total  
537 northern African dust emission is the largest from Region A (west Sahara,  $36\% \pm$   
538  $4.0\%$ ), followed by Region D (central Sahel including the Bodélé Depression,  $21\% \pm$   
539  $4.7\%$ ), Region B (central Sahara,  $13\% \pm 2.6\%$ ), Region C (east Sahara,  $12\% \pm 1.0\%$ ),  
540 and Region E (west Sahel,  $6.5\% \pm 0.64\%$ ). The emission fluxes, however, are the  
541 most intensive in Region D, up to  $11 \pm 2.1 \text{ g month}^{-1} \text{ m}^{-2}$  and are generally below  $5 \text{ g}$   
542  $\text{month}^{-1} \text{ m}^{-2}$  over the other regions.  
543 Concerning the seasonality, higher dust emission tends to occur in boreal spring and  
544 winter, with the largest emission flux of  $19 \pm 4.7 \text{ g month}^{-1} \text{ m}^{-2}$  from Region D. As  
545 shown in Figure 6 and S1, the emissions peak in boreal spring for Region A, B and C,  
546 but in winter for Region D and E. There is also a secondary peak in summer  
547 emissions for Region E. Correlation analysis between dust emissions and  
548 meteorological variables suggests that the seasonality is mainly driven by high surface  
549 wind speeds (with  $r$  of 0.79-0.96 and 0.68-0.97 for the 75<sup>th</sup> and 95<sup>th</sup> percentiles of 10-  
550 m wind speeds, respectively). Apparent negative correlation is also found between  
551 precipitation (soil moisture, Figure S1) and dust emission in Region D with  $r$  of -0.69  
552 (-0.67).  
553 Similar seasonality is also reported by Cowie et al. (2014), who suggested that the  
554 strongest dust season in winter in central Sahel is driven by strong harmattan winds  
555 and frequent Low level Jet breakdown, and the second peak in summer in west Sahel  
556 could be explained by the summer monsoon combined with the Sahara Heat Low. The

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557 study also suggested the dominance of strong wind frequency in the seasonal variation  
558 of the emission frequencies. Fiedler et al. (2013) also found a maximum of emission  
559 flux over the Bodélé Depression in winter and the highest emission flux in spring in  
560 west Sahara. The study suggested that near-surface peak winds associated with  
561 Nocturnal Low-Level Jets is a driver of mineral dust emissions. Negative correlation  
562 between dust emissions and soil moisture has also been revealed by Yu et al. (2017)  
563 and Pierre et al. (2012), as the decreased vegetation growth in response to dry soil  
564 would result in enhanced dust emissions.

565 It is also worth noting that the interannual variation in dust emission is much larger  
566 during the wet season ( $0.96 \pm 0.25 \text{ Pg yr}^{-1}$ , Table 3) than on an annual basis ( $0.73 \pm$   
567  $0.12 \text{ Pg yr}^{-1}$ ). Moreover, while the annual emissions gradually decrease from 2013 to  
568 2017, the emissions during the wet season peak in 2015. The obviously different  
569 behavior between the annual emissions and emissions during the wet season suggests  
570 that predictions of future impact of African dust emissions over the Amazon Basin in  
571 response to climate change should focus on the wet season rather than the annual  
572 average, as the former is more related to the export of African dust towards the  
573 Amazon Basin.

#### 574 **4.2 Transatlantic transport of African dust**

575 The amount of African dust reaching the Amazon Basin depends not only on the dust  
576 emission fluxes, but also the transport path. Associated with the annual oscillation of  
577 ITCZ, the ~~dust column burden shows a steep east-west gradient across the ocean with~~  
578 ~~two major paths for different seasons (Fig. S4): one~~ outflow of African dust moves  
579 slightly southwest toward South America in boreal winter and spring, and ~~the other~~  
580 moves west towards the Caribbean in boreal summer and fall- (Moran-Zuloaga et al.,  
581 2018; Ben-Ami et al., 2012). Therefore, although higher dust load over the coastal  
582 region of North Africa is found in summer ( $> 500 \text{ mg m}^{-2}$ ), dust reaching the Amazon  
583 Basin is less than  $10 \text{ mg m}^{-2}$ . In contrast, dust load over the Amazon Basin could reach  
584 up to  $50 \text{ mg m}^{-2}$  in spring and winter.

585 In addition to the transport path, the changes in dust column burden along the  
586 transport towards the Amazon Basin ~~is~~ are also sensitive to its removal rate, namely  
587 the lifetime against deposition over the Atlantic. Assuming first-order removal of dust

588 aerosols, we further derived seasonal e-folding lifetime (hereinafter referred to as  
589 lifetime,  $\tau$ ) of simulated dust during 2013 – 2017, based on the logarithm of the dust  
590 column burden against travel time over the AOaTP (Fig. 7). Figure 7) using Equation  
591 1:

$$592 \quad \tau = \frac{L}{v \times slope} \quad (1)$$

593 where  $L$  is the distance of 1-degree longitude averaged over 5° S – 25° N in unit of m  
594 degree<sup>-1</sup>;  $v$  is the wind speed in unit of m s<sup>-1</sup>; and  $slope$  is the gradient of the linear  
595 trend line based on the logarithm of dust burden against the distance in degree  
596 between 20 °W and 50 °W.

597 Estimated dust lifetime is the shortest ( $1.4 \pm 0.098$  d) in winter, followed by fall and  
598 spring ( $1.9 \pm 0.33$  d and  $2.3 \pm 0.31$  d, respectively), while the lifetime in summer is  
599 the longest ( $4.2 \pm 0.68$  d). The interannual variability of the lifetime is small in winter  
600 with relative standard deviation (RSD) of 7.0%, but relatively large in fall with RSD  
601 of 17%.

602 The short lifetime in winter is generally associated with high deposition (~~with  $0.18 \pm$~~   
603  ~~$0.034$  Pg a<sup>-1</sup> accounting 20% of the emission of Northern Africa, Table S2). flux~~  
604 ~~(including both dry and wet deposition).~~ As shown in Fig. Figure 8, the spatial  
605 ~~distribution of dust deposition is similar to that of dust burden, again illustrating the~~  
606 ~~main transport paths. The~~ largest dust deposition flux ( $> 1000$  ng m<sup>-2</sup> s<sup>-1</sup>) is found over  
607 the source regions in northern Africa, especially in spring and winter, and is mainly  
608 due to dry deposition (accounting for more than 80%). As a result, 48% – 64% of total  
609 emission in northern Africa is deposited within the source region (Table S2). The  
610 deposition flux over the AOaTP, also shows strong seasonality, with a maximum of  
611  $\sim 530$  ng m<sup>-2</sup> s<sup>-1</sup> in winter and a minimum of  $\sim 180$  ng m<sup>-2</sup> s<sup>-1</sup> in fall, and is mainly  
612 driven by wet deposition (accounting for 76% on average). ~~Again, although the~~  
613 ~~emissions are similar in spring and winter, the deposition flux is much larger in~~  
614 ~~winter, consistent with the relatively shorter lifetime in winter discussed above. On~~  
615 ~~the other hand, the highest dust burden ( $144 \pm 58$  mg m<sup>-2</sup>) over the AOaTP is found in~~  
616 ~~summer mainly associated with its longer lifetime, followed by  $127 \pm 24$ ,  $98 \pm 35$ , and~~

617 ~~57 ± 20 mg m<sup>-2</sup> in winter, spring and fall, respectively.~~ The deposition over the  
618 AOA<sub>TP</sub> ~~only~~ accounts for ~~7.720%~~ of total emission in northern Africa in ~~Spring~~winter,  
619 in contrast to ~~207.7%~~ in ~~spring, consistent with the relatively shorter lifetime~~ in  
620 winter. ~~— discussed above.~~

621 ~~5. The seasonality in the deposition fluxes and the consequent dust lifetime depends~~  
622 ~~not only on precipitation but also the vertical pathways of dust transport across the~~  
623 ~~Atlantic. Dust aerosols aloft at higher altitude reach further west and have relatively~~  
624 ~~longer lifetime. Significant differences in dust vertical distributions along the~~  
625 ~~transport pathways have been revealed from the CALIOP measurements, which show~~  
626 ~~that more dust is transported above 2km in summer while the dust layer is the~~  
627 ~~shallowest in winter (Liu et al., 2012).~~

#### 628 **4.3 The influence of African dust over the Amazon Basin**

##### 629 **4.3.1 Surface aerosol concentrations and AOD**

630 Figure 9 shows the time series of observed and simulated aerosol mass concentrations  
631 at ATTO in January – June for the period of 2014 – 2016. Observed mean concentration  
632 in wet season is  $9.3 \pm 7.6 \mu\text{g m}^{-3}$ , of which 83% is from coarse aerosol ( $7.7 \mu\text{g m}^{-3}$ ),  
633 while simulated concentration is  $11 \pm 6.7 \mu\text{g m}^{-3}$ , with dust contribution of 65% ( $7.2 \mu\text{g m}^{-3}$ ).  
634 The slight model bias could be to some extent explained by the difference in  
635 background concentrations ( $1.9$  and  $5.1 \mu\text{g m}^{-3}$  for the observation and model data,  
636 respectively). ~~During the wet season, observed coarse aerosol concentrations frequently~~  
637 ~~exceed  $9 \mu\text{g m}^{-3}$ , and could be up to  $50 \mu\text{g m}^{-3}$ .~~ Most of observed peaks are found in  
638 February – March of 2014 and 2016, and in February – April of 2015. The high  
639 correlation ( $r$  of  $0.52 - 0.71$ ) between observed coarse aerosols and simulated dust  
640 concentrations suggests that observed strong variation in coarse aerosols is mainly  
641 driven by the influence of dust. ~~Rizzolo et al. (2017) conducted aerosol measurements~~  
642 ~~at ATTO from 19 March to 24 April 2015. The study showed the arrival of African dust~~  
643 ~~between 3 and 6 April when the highest concentrations of PM<sub>10</sub>, soluble Fe (III) and Fe~~  
644 ~~(II) were recorded at ATTO. The peak value of  $23 \mu\text{g m}^{-3}$  for PM<sub>10</sub> was observed on 5~~  
645 ~~April. This dust event is well reproduced in this study with the peak value of  $28 \mu\text{g m}^{-3}$~~   
646 ~~for PM<sub>10</sub> on the same day and the dust contribution to PM<sub>10</sub> reaching above 70%. The~~  
647 ~~co-occurrence of elevated sea salt concentration (reaching  $2.5 \mu\text{g m}^{-3}$ ) during this event~~  
648 ~~is also found in this study, consistent with previous studies which show mixed transport~~



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649 [of African dust and marine aerosol to the basin \(Wang et al., 2016; Ben-Ami et al., 2010;](#)  
650 [Rizzolo et al., 2017; Adachi et al., 2020\).](#)

651 The dust peaks are generally associated with large dust emission and/or efficient  
652 trans-Atlantic transport (e.g. relatively longer lifetime). For example, the relatively  
653 higher dust concentrations in the wet season of 2015 (except for February) are  
654 generally associated with higher emissions ( $1.2 - 1.5 \text{ Pg } \text{ayr}^{-1}$ ) compared with the  
655 year of 2014 and 2016 ( $0.68 - 1.0 \text{ Pg } \text{ayr}^{-1}$ , see Table S3). On the other hand, although  
656 emissions in February 2016 ( $0.95 \text{ Pg } \text{ayr}^{-1}$ ) is slightly lower than those in February  
657 2014 ( $1.2 \text{ Pg } \text{ayr}^{-1}$ ), the relatively longer lifetime (1.7 d vs. 1.5 d) may help explain  
658 the high dust concentrations during that period. It should be noted that the lifetime  
659 estimated here represents the export efficiency averaged over a relatively large  
660 domain and a long-time scale (e.g. one month). Besides, the influence of African dust  
661 on the ATTO site is also subject to the variations of transport paths and precipitation  
662 fields.

663 Over the whole Amazon Basin, simulated [average mean](#) surface dust concentrations in  
664 the wet season of 2013 – 2017 are  $5.7 \pm 1.3 \mu\text{g m}^{-3}$ , with ~~maxima over 15~~ [a maximum](#)  
665 [of  \$7.9 \mu\text{g m}^{-3}\$  in 2015 driven mainly by the relatively high dust emission flux. The](#)  
666 [maxima of surface dust concentrations are found in](#) the northeast corner of rainforest  
667 ~~and a~~ [\(over  \$15 \mu\text{g m}^{-3}\$ \) with a clearly](#) decreasing trend towards southwestern direction  
668 ~~(Fig. Figure 10). The dust contribution to surface aerosol concentrations is  $47\% \pm$~~   
669  ~~$5.0\%$  (up to  $70\%$  in the north corner); averaged over the whole basin is  $40\% \pm 4.5\%$ ,~~  
670 [again with the maximum of  \$48\%\$  found in 2015.](#) The location with the largest dust  
671 contributions [\(up to  \$70\%\$  in the north corner\)](#) slightly shifted inland compared to the  
672 spatial distribution of dust concentration. This could be explained by [relatively](#) higher  
673 influence of sea salt aerosols along the coast. ~~The ATTO site has dust concentrations~~  
674 ~~(around  $8.1 \pm 1.8 \mu\text{g m}^{-3}$  (accounting for  $63\% \pm 7.9\%$  of total aerosol concentrations~~  
675 ~~on average) in wet season, and thus could be representative of the whole Amazon~~  
676 ~~Basin. Based on single-particle analysis using a quantitative energy-dispersive~~  
677 ~~electron probe X-ray microanalysis, Wu et al. (2019) found that aged mineral dust and~~  
678 ~~sea salts account for  $37\% - 70\%$  of the super-micron aerosol at ATTO site during the~~

wet season, consistent with our result. The contribution 30-50% near the coast of ~~DOD~~ to AOD at 675 nm over most areas of the Amazon Basin (Fig. 11) is in South America). Although the range of 10 – 50% (26% ± 4.7% on average) during the wet season of 2013 – 2017, with maxima in the northern Amazon Basin. The emission fluxes of both sea salt and dust contribution to total AOD is relatively smaller than that to surface aerosol concentrations, mainly because of the relatively lower MEE are largely determined by surface winds, the interannual variability of dust aerosols compared to other aerosols.

**5.2 Frequency of dust events** concentrations is larger than sea salt over the Amazon Basin (20% vs. 10%) as the former is also sensitive to the export efficiency across the Atlantic Ocean as discussed above.

Figure 10c also shows the frequency of dust events over the Amazon Basin, estimated as the number of days when daily surface dust concentrations ~~reach~~ reaching the threshold of  $9 \mu\text{g m}^{-3}$  ~~on daily basis as defined in~~ (Moran-Zuloaga et al. (., 2018) ~~over~~ divided by the Amazon Basin total number of days in the wet season of 2013 – 2017. Dust frequency averaged over the whole region is around  $18\% \pm 4.6\%$  and decreases from 50 – 60% at the northeast coast to  $< 1\%$  in southern inland. The frequency of dust events at ATTO site is around 32%, close to the median of the range. The interannual variation of the frequency, (represented by RSD), however, has an opposite trend, ~~with RSD~~ gradually increasing from 10% at the northeast coast to over 100% in southern inland (36% at ATTO). During dust events, the dust mass concentration ~~of~~ at ATTO reaches  $16 \pm 2.9 \mu\text{g m}^{-3}$  (three times as high as that over the whole wet season), accounting for around ~~77.5%~~  $\pm 5.83\%$  of total aerosol. (Figure 10d). Similarly, under the influence of the long-range transport of Saharan dust plumes, Moran-Zuloaga et al. (2018) observed mass concentrations  $14 \pm 12 \mu\text{g m}^{-3}$  for coarse aerosol at the same site, accounting for 93% of total observed aerosol.

### **4.3.2 AOD**

The contribution of DOD to AOD at 550 nm over most areas of the Amazon Basin

(Figure 11) is in the range of 10 – 50% ( $23\% \pm 4.4\%$  on average) during the wet season of 2013 – 2017, with maxima in the northern Amazon Basin. The dust contribution to total AOD is relatively smaller than that to surface aerosol concentrations, mainly because of the relatively lower MEE of dust aerosols compared to other aerosols. There also exists large difference in DOD between the whole wet season and dust events:  $0.019021 \pm 0.00470055$  vs.  $0.038055 \pm 0.0074$  (at wavelength of 675 nm)  $0.0076$  averaged over the Amazon Basin. A maximum of 0.31 on a daily basis is found on 1 Mar 2016 at the northeast corner ( $55^\circ$  W,  $4^\circ$  N) of the Amazon Basin during the study period. During dust events, dust aerosols dominate AOD ( $50.40\% - 60\%$ ) over most regions of the Amazon Basin. At ATTO site, DOD is  $0.029034 \pm 0.00760088$  and  $0.054063 \pm 0.00740087$ , accounting for  $41.37\%$  and  $57.53\%$  of AOD over the whole wet season and dust events, respectively. The largest dust contribution (up to  $84.81\%$ ) with DOD of  $0.1518$  at ATTO site is found on 24 Jan 2015. Consistent with our results, previous studies by Baars et al. (2011) and Baars et al. (2012) reported DOD (532 nm) of up to 0.18 and AOD of  $\sim 0.14$  when affected by strong influence of Saharan dust at a similar Amazon site ( $60^\circ 2.3'$  W,  $2^\circ 35.9'$  S).

### 5.4.3.3 Dust deposition and related nutrient input

The spatial distribution of dust deposition over the Amazon Basin is also shown in Fig. Figure 8. The mean dust deposition flux in wet season is  $2.0 \pm 0.35 \text{ g m}^{-2} \text{ yr}^{-1}$ , much higher than in dry seasons (August to November,  $0.35 \pm 0.16 \text{ g m}^{-2} \text{ yr}^{-1}$ ). The maximum ( $2.6 \text{ g m}^{-2} \text{ yr}^{-1}$ ) is found in the year 2015 due to relatively large dust emission and efficient trans-Atlantic transport. With emission of  $0.96 \pm 0.25 \text{ Pg yr}^{-1}$  in wet season ( $0.73 \pm 0.12 \text{ Pg yr}^{-1}$  on annual average), only 1.9% (1.4%) of African dust is deposited into the Amazon Basin (dominated by wet deposition) while relatively large part is deposited over the AOaTP (13% in the wet season and 14% on annual average) and northern Africa (49% in the wet season). Assuming mass fractions of 4.4%, 0.082%, and 1.8% for iron, phosphorus, and magnesium respectively in the African dust (Bristow et al., 2010; Chiemeka et al.,

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737 2007), we derive deposition fluxes of  $88 \pm 15 \text{ mg m}^{-2} \text{ yr}^{-1}$ ,  $1.6 \pm 0.29 \text{ mg m}^{-2} \text{ yr}^{-1}$  and  
738  $36 \pm 6.3 \text{ mg m}^{-2} \text{ yr}^{-1}$  for iron, phosphorus and magnesium respectively into the  
739 Amazon rainforest during the wet season and  $52 \pm 8.7$ ,  $0.97 \pm 0.16$  and  $21 \pm 3.6 \text{ mg}$   
740  $\text{m}^{-2} \text{ yr}^{-1}$  on annual average (Fig.Figure 12). It should be noted that there exists large  
741 spatial variation of nutrient input into the Amazon Basin associated with the patterns  
742 of dust burden and dust deposition flux. The deposition flux decreases from over  $70$   
743  $\text{mg m}^{-2} \text{ yr}^{-1}$  at northeast coast to less than  $7 \text{ mg m}^{-2} \text{ yr}^{-1}$  in inland for magnesium  
744 and decreases from  $> 9 \text{ mg m}^{-2} \text{ yr}^{-1}$  at northeast coast to less than  $1 \text{ mg m}^{-2} \text{ yr}^{-1}$  in  
745 southwestern Basin for phosphorus during the wet season. Similarly, the deposition  
746 flux of iron during the wet season decreases from over  $500 \text{ mg m}^{-2} \text{ yr}^{-1}$  at northeast  
747 coast to less than  $15 \text{ mg m}^{-2} \text{ yr}^{-1}$  in the southwest and is above  $50 \text{ mg m}^{-2} \text{ yr}^{-1}$  in  
748 most of the Amazon Basin. It seems that the nutrient input from Africa dust may play  
749 a significant role in the northeastern part of the Amazon Basin, not in the southwest.  
750 Table 34 summarized the estimates of dust and associated phosphorus deposition into  
751 the Amazon Basin from previous studies. The estimated fluxes of dust and associated  
752 phosphorus deposition are in the range of  $0.81 - 19 \text{ g m}^{-2} \text{ yr}^{-1}$  and  $0.48 - 16 \text{ mg m}^{-2}$   
753  $\text{yr}^{-1}$ . The large range is mainly driven by the high values ( $19 \text{ g m}^{-2} \text{ yr}^{-1}$  and  $16 \text{ mg m}^{-2}$   
754  $\text{yr}^{-1}$  for dust and associated phosphorus, respectively) from the study of Swap et al.  
755 (1992). Based on observations during storm events and dust climatology, the study  
756 estimated dust importation into the northeastern basin, which is most subject to the  
757 intrusion of African dust. Besides the discrepancy in defined regions, the wide range  
758 could also be partly explained by the application of different methods and associated  
759 intrinsic uncertainties as mentioned in the Introduction. For instance, the estimates  
760 from Swap et al. (1992) are mainly based on 1-month field measurements at three  
761 sites located in the northeastern basin. Assumption about air exchange rate across the  
762 coast to the basin, duration of dust storms as well as dust concentrations contained in  
763 the dust plumes had to be made to extrapolate the dust deposition into the Amazon  
764 Basin. Similarly, bias could also arise from insufficient observations available to  
765 constrain models or satellite retrievals. Additional uncertainty may also stem from the

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766 assumption about the P mass fraction, ranging from 0.07% to 0.108%. Our results are  
767 similar to the finding of Prospero et al. (2020), which has also been constrained by the  
768 observation at Cayenne. \_

769 According to Salati and Vose (1984), the total amount of phosphorous and magnesium  
770 is  $21.6 \text{ g m}^{-2}$  and  $29.8 \text{ g m}^{-2}$ , respectively, in the ecosystem of the Amazon Basin ( $14.7$   
771 and  $2.3 \text{ g m}^{-2}$  respectively in the soil). On the other hand, Vitousek and R-L-Sanford  
772 (1986) reported a loss of  $0.8 - 4 \text{ mg m}^{-2} \text{ a}^{-1}$  for phosphorus and  $810 \text{ mg m}^{-2} \text{ a}^{-1}$  for  
773 magnesium in Brazilian ecosystem to surface waters. Estimated nutrient input from  
774 African dust in our study accounts for 0.011% and 1.6% of total phosphorous and  
775 magnesium in the soil over the Amazon Basin during the wet season (0.0066% and  
776 0.91% on annual average), and could almost compensate the hydrologic losses of  
777 phosphorous in Brazilian forest ecosystem. Similarly, Abouchami et al. (2013)  
778 pointed out that most of the Amazonian rainforest is a system with an internal  
779 recycling of nutrients. But the extra influx of nutrients from African dust might  
780 account for a significant portion of the net outflux, i.e. dissolved discharge of nutrients  
781 into rivers. Keep in mind that the estimates of nutrients influx are subject to the  
782 uncertainties in the estimates of dust flux as well as the mass fractions of nutrients  
783 contained in the dust. In addition, marine aerosols and biomassburning aerosols  
784 mixed with the LRT of African dust may also play a role for certain essential nutrients  
785 (Prospero et al., 2020; Abouchami et al., 2013). More observations including the mass  
786 fraction of nutrient in dust aerosols and the deposition fluxes of those elements in the  
787 Amazon Basin are necessarily required in the future work to better evaluate the  
788 nutrient input associated with the African dust intrusion.

789

## 790 5 ~~6~~-Conclusion

791 In this study, we use the GEOS-Chem model with optimized particle mass size  
792 distribution (PMSD) of dust aerosols to investigate the influence of the export of  
793 African dust towards the Amazon Basin during 2013 – 2017. The model performance  
794 is constrained by multiple datasets obtained from AERONET, MODIS, as well as

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795 Cayenne and ATTO sites in the Amazon Basin, ~~including particle size distribution~~  
796 ~~over Africa, aerosol optical depth (AOD) over Africa and the Atlantic Ocean as well~~  
797 ~~as coarse and total aerosols concentrations in the Amazon Basin.~~  
798 ~~Simulated dust emission from northern Africa is  $0.73 \pm 0.12 \text{ Pg a}^{-1}$ , accounting for~~  
799 ~~more than 70% of global dust emission. There exists a strong seasonality in dust~~  
800 ~~emission with peaks in spring or winter, which varies with source regions and is~~  
801 ~~mainly driven by high surface wind speeds. It is worth noting that no consistent~~  
802 ~~decline is found for dust emission during the wet season, when the export of African~~  
803 ~~dust towards the Amazon Basin is more efficient driven by the southward movement~~  
804 ~~of ITCZ.~~  
805 ~~In addition to the transport path associated with the oscillation of ITCZ, the export~~  
806 ~~efficiency of African dust towards the Amazon basin is sensitive to the removal of~~  
807 ~~dust aerosol along the trans-Atlantic transport, which also depends on assumed PMSD~~  
808 ~~of dust aerosols in the model.~~ The optimized PMSD in this study well captures  
809 observed AOD regarding both the mean value as well as the decline rate of the  
810 logarithm of AOD over the Atlantic Ocean along the transport path (AOaTP), while  
811 the other two PMSD schemes tend to overestimate the decline rate by 11% and  
812 underestimate the mean value by up to ~40%.

813 Simulated dust emission from northern Africa is  $0.73 \pm 0.12 \text{ Pg yr}^{-1}$ , accounting for  
814 more than 70% of global dust emission. There exists a strong seasonality in dust  
815 emission with peaks in spring or winter, which varies with source regions. The  
816 correlation analysis suggests high surface wind speeds and low soil moisture as a  
817 major driver for dust emissions. In addition to the transport path associated with the  
818 oscillation of ITCZ, the export efficiency of African dust towards the Amazon basin is  
819 sensitive to the removal of dust aerosol along the trans-Atlantic transport, which  
820 depends on both assumed PMSD of dust aerosols in the model and meteorological  
821 fields (i.e. precipitation and vertical transport path). The study further estimates the e-  
822 folding lifetime of dust aerosols along the trans-Atlantic transport based on the  
823 logarithm of the dust column burden against travel time over the AOaTP. The shortest

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824 lifetime (1.4 d) is found for winter associated with high deposition flux, while the  
825 highest dust burden over the AOaTP is found in summer mainly associated with its  
826 longer lifetime (4.2 d).  
827 Simulated surface dust concentration averaged over the whole Amazon Basin is  $5.7 \pm$   
828  $1.3 \mu\text{g m}^{-3}$  during the wet season of 2013 – 2017, contributing  $47\% \pm 40\% \pm 4.5\%$   
829 to total surface aerosols. Observed dust peaks at the ATTO site are generally  
830 associated with large dust emission and/or efficient trans-Atlantic transport. The  
831 frequency of dust events is  $18\% \pm 4.6\%$  averaged over the Amazon Basin and up to  
832  $50\% - 60\%$  at the northeast coast. During the dust events, DOD is around  $0.038055 \pm$   
833  $0.0076$  and dominate total AOD over most of the Amazon Basin. Associated with the  
834 deposition of African dust, the study estimated annual inputs of  $52 \pm 8.7$ ,  $0.97 \pm 0.16$   
835 and  $21 \pm 3.6 \text{ mg m}^{-2} \text{ a}^{-1}$  for iron, phosphorus and magnesium into the Amazon  
836 rainforest, which may well compensate the hydrologic losses of nutrients in the forest  
837 ecosystem.

838

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865

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869

870 Data availability.

871 OPS data used in this study could be found at <https://www.attodata.org/>. Other datasets are  
872 available upon request.

873

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**Table 1.** ~~<sup>1</sup>) in northern Africa (17.5° W – 40° E, 10° N – 35° N)<sup>a</sup>:~~

<del>Year</del>	<del>Spring</del>	<del>Summer</del>	<del>Fall</del>	<del>Winter</del>	<del>Annual (Wet season)</del>
<del>2013</del>	<del>1.2</del>	<del>0.77</del>	<del>0.48</del>	<del>1.0</del>	<del>0.88 (1.1)</del>
<del>2014</del>	<del>0.83</del>	<del>0.84</del>	<del>0.51</del>	<del>0.91</del>	<del>0.77 (0.89)</del>
<del>2015</del>	<del>1.2</del>	<del>0.46</del>	<del>0.33</del>	<del>1.1</del>	<del>0.77 (1.3)</del>
<del>2016</del>	<del>0.82</del>	<del>0.52</del>	<del>0.37</del>	<del>0.89</del>	<del>0.65 (0.86)</del>
<del>2017</del>	<del>0.68</del>	<del>0.38</del>	<del>0.47</del>	<del>0.70</del>	<del>0.56 (0.63)</del>
<del>Mean ± std<sup>b</sup></del>	<del>0.95 ± 0.24</del>	<del>0.59 ± 0.20</del>	<del>0.43 ± 0.078</del>	<del>0.92 ± 0.15</del>	<del>0.73 ± 0.12 (0.96 ± 0.25)</del>

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~~<sup>a</sup> Spring: March – May; Summer: June – August; Fall: September – November; Winter: January;~~

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~~February, and December; Wet season: January – April~~

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~~<sup>b</sup> standard deviation~~

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**Table 2.** Mass fractions (%) of dust emitted in each bin for different particle mass size distribution (PMSD) schemes tested in GEOS-Chem.

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Scheme	bin 1				bin 2	bin 3	bin 4
	sub-bin 1	sub-bin 2	sub-bin 3	sub-bin 4			
	(0.1 – 0.18) <sup>a</sup>	(0.18 – 0.3) <sup>a</sup>	(0.3 – 0.6) <sup>a</sup>	(0.6 – 1.0) <sup>a</sup>			
	(3.1) <sup>b</sup>	(4.3) <sup>b</sup>	(2.7) <sup>b</sup>	(0.96) <sup>b</sup>	(0.45) <sup>b</sup>	(0.27) <sup>b</sup>	(0.16) <sup>b</sup>
V12	0.7	3.32	24.87	71.11	19.2	34.9	38.2
V12_C	6	12	24	58.00	25.3	32.2	30.2
V12_F	3.9	8.06	43	45.04	11.9	15.6	67

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<sup>a</sup> size range in radius (μm) for each bin

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<sup>b</sup> mass extinction efficiency (MEE) at wavelength of 550 nm in unit of m<sup>2</sup> g<sup>-1</sup> for dust particles in

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each bin in the GEOS-Chem ~~model~~mod

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**Table 2.** Summary of the observations used in this study, including the parameters,

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the spatio-temporal coverage, and the corresponding application in the model.

<u>Datasets</u>	<u>Parameters</u>	<u>Locations</u>	<u>Periods</u> <u>(resolution)</u>	<u>Application</u>
<u>AERONET</u>	<u>AOD</u>	<u>northern Africa, the</u> <u>Atlantic Ocean</u>	<u>2013 – 2017</u> <u>(daily)</u>	<u>Model AOD evaluation over northern</u> <u>Africa and the Atlantic Ocean</u>

	<u>PVSD<sup>a</sup></u>	<u>northern Africa</u>	<u>2013 – 2017</u> <u>(daily)</u>	<u>Model dust PMSD evaluation</u>
<u>Fennec</u> <u>Campaign</u>	<u>PMSD<sup>b</sup></u>	<u>Over Mali and</u> <u>Algeria, Africa</u>	<u>17 – 28 June,</u> <u>2011</u>	<u>Model dust PMSD evaluation</u>
<u>MODIS</u>	<u>AOD</u>	<u>northern Africa and</u> <u>the Atlantic Ocean</u>	<u>2013 – 2017</u> <u>(daily)</u>	<u>Model AOD evaluation over northern</u> <u>Africa and the Atlantic Ocean</u>
<u>Cayenne</u>	<u>PM<sub>10</sub></u>	<u>52.3097° W,</u> <u>4.9489° N (France)</u>	<u>January – April,</u> <u>2014 (daily)</u>	<u>Model dust mass concentration</u> <u>evaluation at the coast of South America</u>
<u>ATTO</u>	<u>PNSD<sup>c</sup></u>	<u>59.0056° W, 2.1459°</u> <u>S (Brazil)</u>	<u>January – April,</u> <u>2014 – 2016</u> <u>(5 min)</u>	<u>Model dust mass concentration</u> <u>evaluation at the central Amazon Basin</u>

1285 <sup>a</sup> particle volume size distribution; <sup>b</sup> particle mass size distribution; <sup>c</sup> particle number size  
1286 distribution

1287 **Table 3.** Annual and seasonal dust emissions (Pg yr<sup>-1</sup>) in northern Africa (17.5° W –  
1288 40° E, 10° N – 35° N)<sup>a</sup>.

<u>Year</u>	<u>Spring</u>	<u>Summer</u>	<u>Fall</u>	<u>Winter</u>	<u>Annual (Wet season)</u>
<u>2013</u>	<u>1.2</u>	<u>0.77</u>	<u>0.48</u>	<u>1.0</u>	<u>0.88 (1.1)</u>
<u>2014</u>	<u>0.83</u>	<u>0.84</u>	<u>0.51</u>	<u>0.91</u>	<u>0.77 (0.89)</u>
<u>2015</u>	<u>1.2</u>	<u>0.46</u>	<u>0.33</u>	<u>1.1</u>	<u>0.77 (1.3)</u>
<u>2016</u>	<u>0.82</u>	<u>0.52</u>	<u>0.37</u>	<u>0.89</u>	<u>0.65 (0.86)</u>
<u>2017</u>	<u>0.68</u>	<u>0.38</u>	<u>0.47</u>	<u>0.70</u>	<u>0.56 (0.63)</u>
<u>Mean±std<sup>b</sup></u>	<u>0.95±0.24</u>	<u>0.59±0.20</u>	<u>0.43±0.078</u>	<u>0.92±0.15</u>	<u>0.73±0.12 (0.96±0.25)</u>

1289 <sup>a</sup> Spring: March – May; Summer: June – August; Fall: September – November; Winter: January,  
1290 February, and December; Wet season: January – April

1291 <sup>b</sup> standard deviation

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1300 **Table 34.** Estimates of annual dust and associated phosphorus deposition into the  
1301 Amazon Basin.

Methods	Dust deposition		P deposition		References
	total (Tg a <sup>-1</sup> )	flux (g m <sup>-2</sup> a <sup>-1</sup> )	total (Tg a <sup>-1</sup> )	flux (mg m <sup>-2</sup> a <sup>-1</sup> )	
CESM2	10 ± 2.1	n/a	0.0077±0.0016	n/a	Li et al. (2021) <sup>a</sup>
AeroCom Phase I	7.7	0.81	0.0063	0.66	Kok et al. (2021) <sup>b</sup>
MERRA-2	8.0	1.05	0.0062	0.9	Prospero et al. (2020) <sup>a</sup>
MERRA-2, CAM	n/a	n/a	0.011 – 0.033	1.1 – 3.5	Barkley et al. (2019) <sup>a</sup>
GLOMAP	32	1.8	0.019	1.1	Herbert et al. (2018) <sup>a</sup>
CALIOP	8– 48	0.8 – 5	0.006 – 0.037	0.7 – 3.9	Yu et al. (2015b) <sup>a</sup>
ECHAM5	30.3/11.4	n/a	0.025/0.0093	n/a	Gläser et al. (2015) <sup>b</sup>
GEOS-Chem	17± 5	n/a	0.014	n/a	Ridley et al. (2012) <sup>b</sup>
MATCH	n/a	n/a	n/a	0.48	Mahowald et al. (2005) <sup>a</sup>
MODIS	50	n/a	0.041	n/a	Kaufman (2005) <sup>b</sup>
Field measurement	13	19	0.011	16	Swap et al. (1992) <sup>b</sup>
GEOS-Chem	10 ± 1.7	1.2 ± 0.20	0.0085 ± 0.0014	0.97 ± 0.16	This study

1302 *Note.* Table extracted in part from Prospero et al. (2020).

1303 <sup>a</sup> The P mass fraction is 0.077% for Li et al. (2021) and Prospero et al. (2020), 0.108% for Barkley  
1304 et al. (2019), 0.088% for Herbert et al. (2018), 0.078% for Yu et al. (2015b), and 0.07% for  
1305 Mahowald et al. (2005).

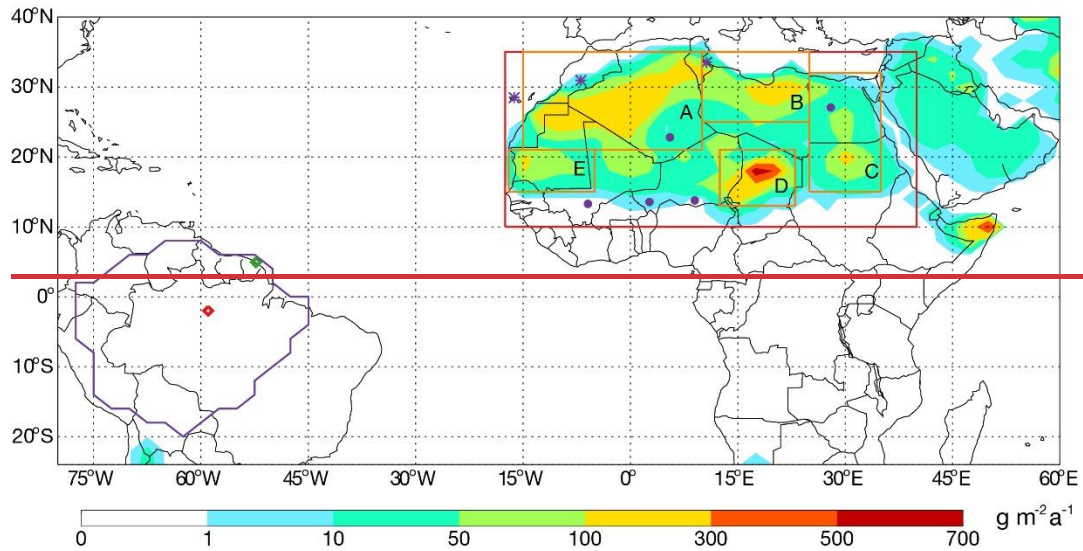
1306 <sup>b</sup> Assuming P mass fraction of 0.082% in dust, the same value as used in this study.

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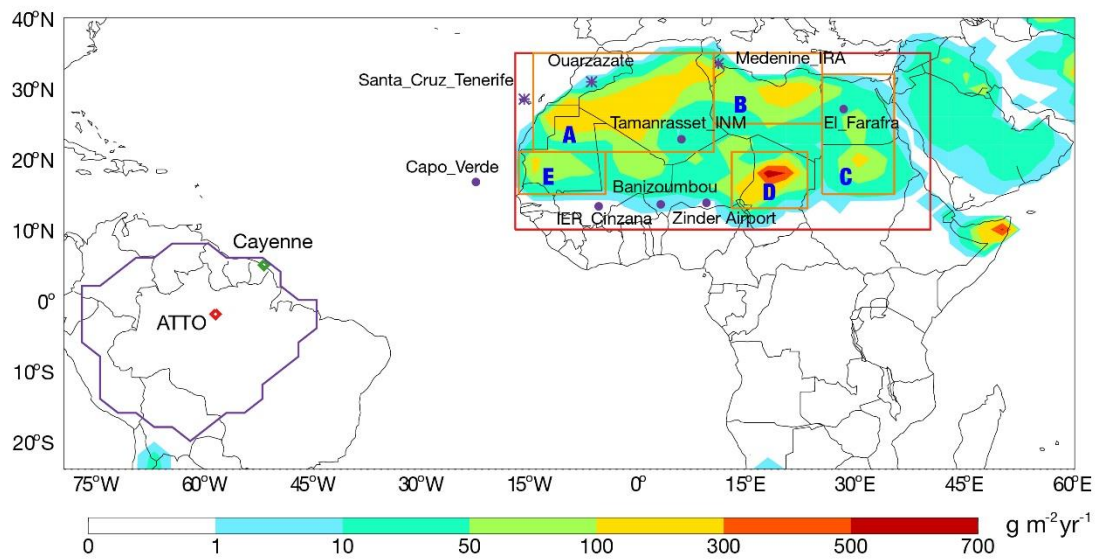
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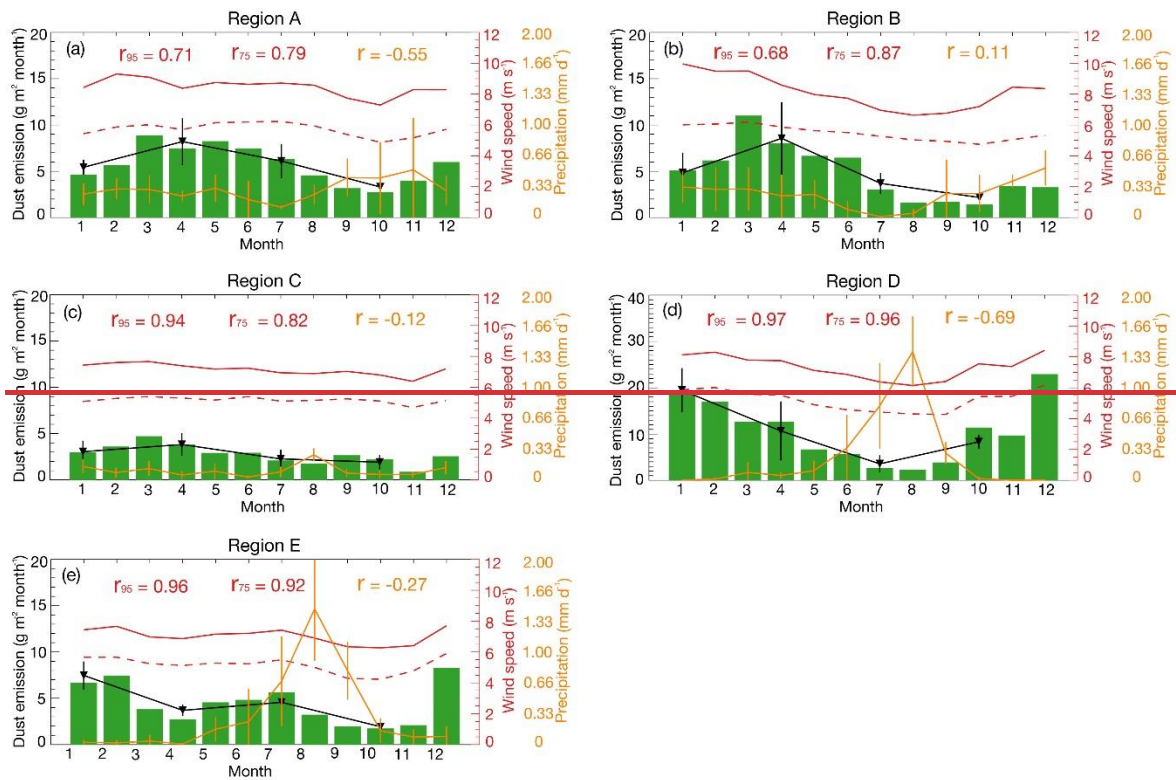
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**Figure 1.** Simulated annual dust emissions in GEOS-Chem, averaged from 2013 to 2017. The location of AERONET sites used in [Fig. 3](#) [Figure 2](#) are marked as purple symbols, of which circles represent the sites used in [Fig. 4](#) [Figure 3](#). The region of the Amazon Basin is defined by purple lines. The location of Cayenne site in the northeast coast of South America and ATTO site in the central Amazon Basin are marked as green and red diamonds, respectively. The red rectangle illustrates the area of northern Africa (17.5° W – 40° E, 10° N – 35° N) and the orange rectangles shows the areas of five major source regions described in the text (A: 15° W – 10° E, 21° N – 35° N; B: 10° E – 25° E, 25° N – 35° N; C: 25° E – 35° E, 15° N – 32° N; D: 12.5° E – 23° E, 13° N – 21° N; E: 17° W – 5° W, 15° N – 21° N).—





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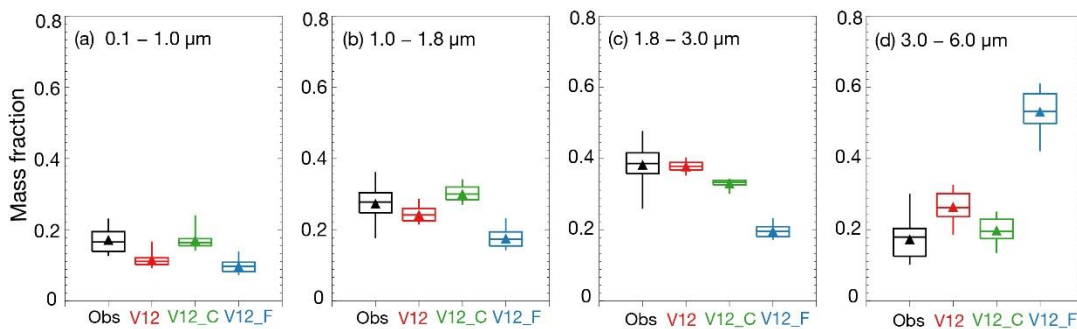
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**Figure 2.** Monthly dust emission fluxes together with the 95<sup>th</sup> percentile hourly wind speeds (red solid lines), the 75<sup>th</sup> percentile hourly wind speeds (red dotted lines) and precipitation (yellow lines) over the five major source regions averaged from 2013 to 2017. Seasonal emission fluxes of dust are also shown as black lines. The correlation coefficients (r) between the dust emission fluxes and different meteorological variables are also shown in each panel.

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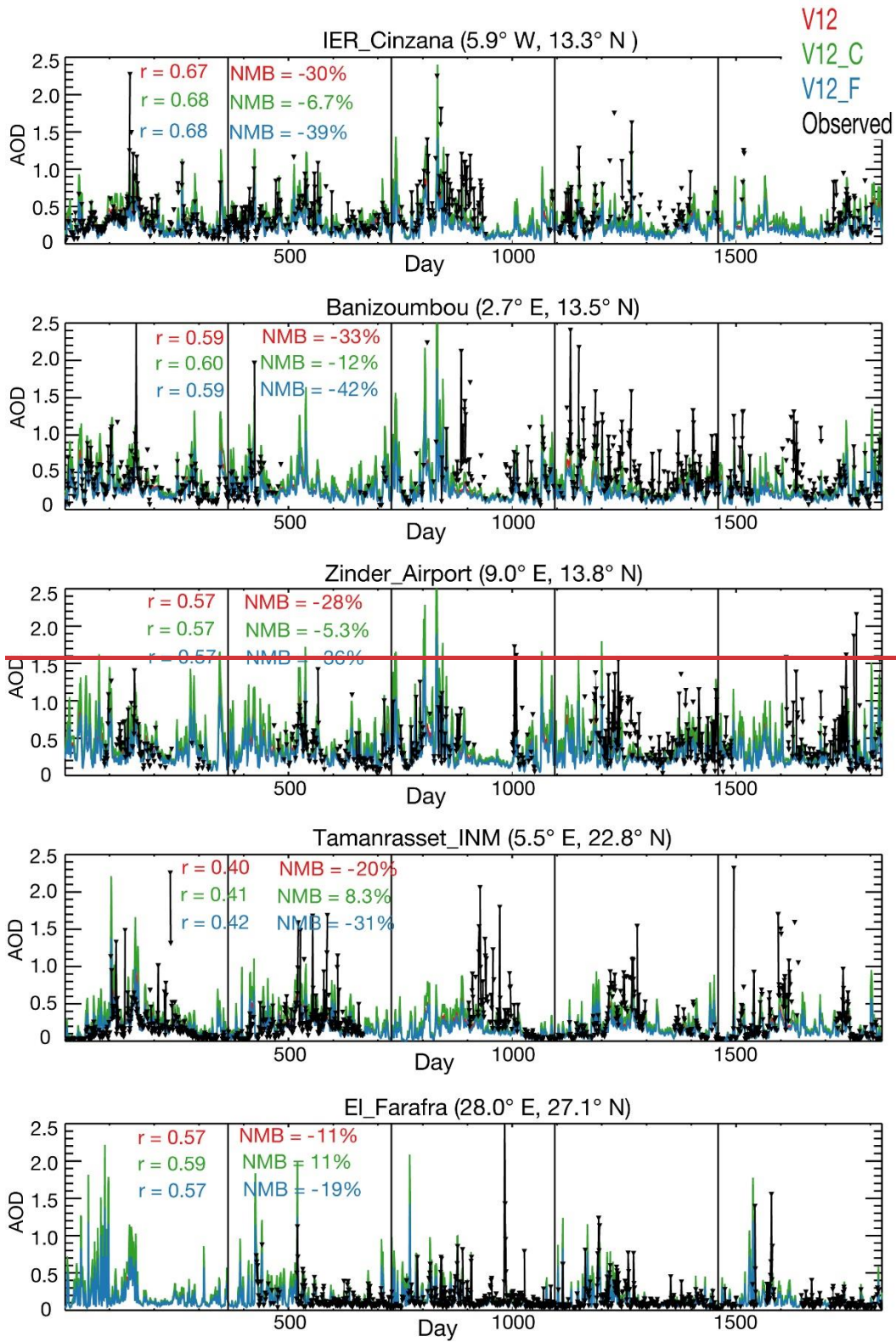


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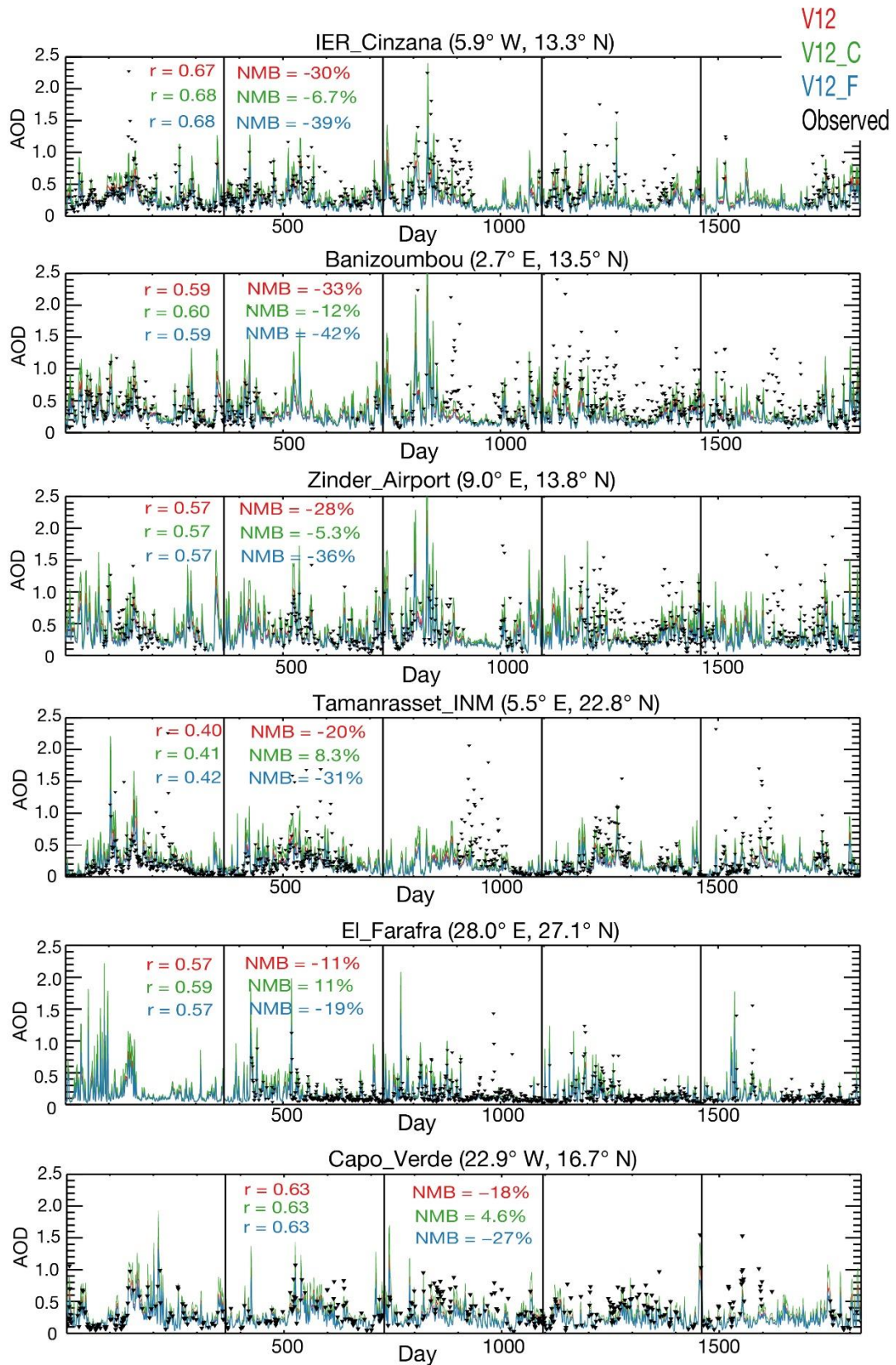
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**Figure 32.** Boxplots of the mass fractions of column-integrated aerosols in the four size bins (in radius) retrieved from AERONET sites over Africa compared with model results based on different PMSD schemes. The triangles represent the mean values.



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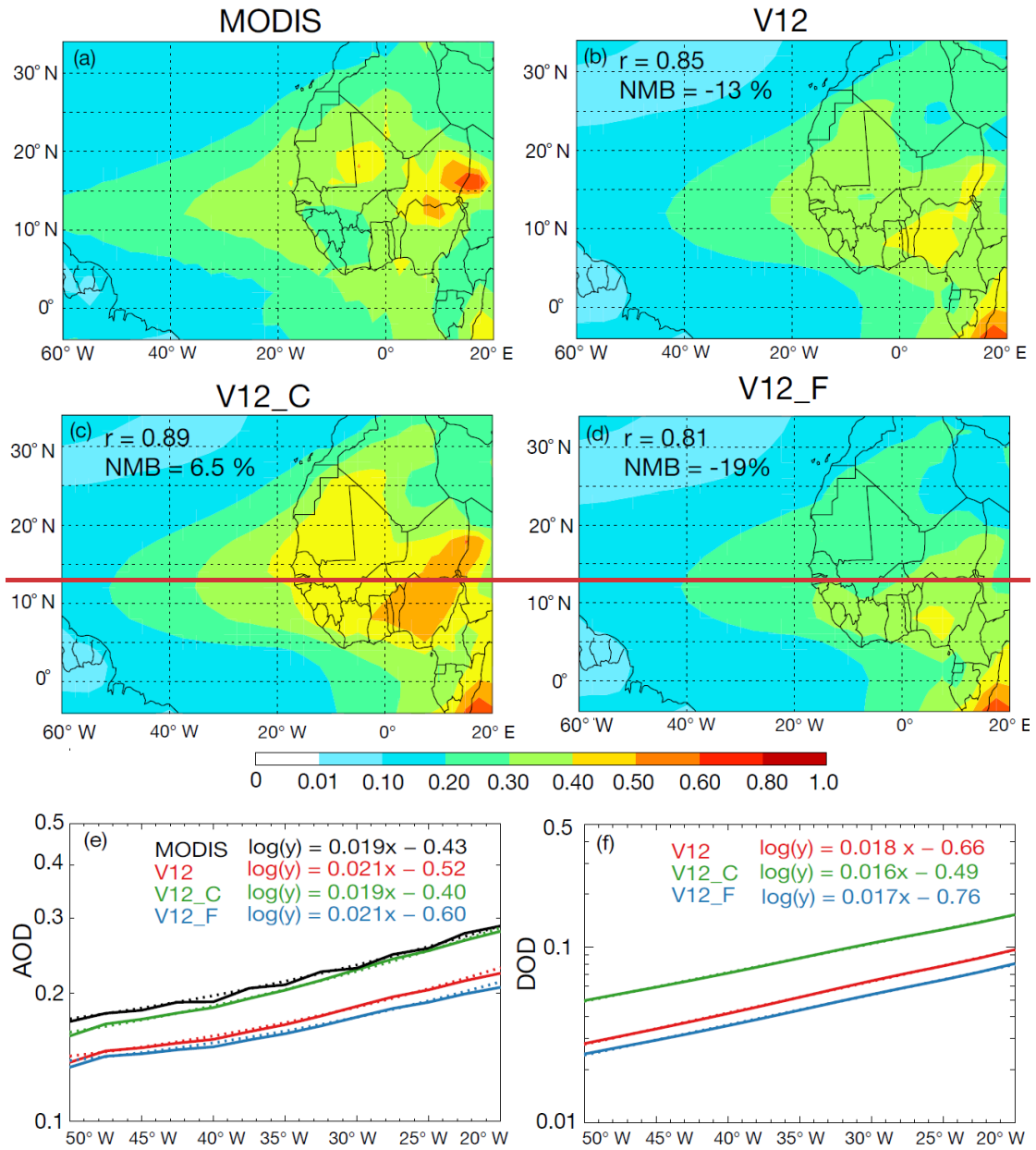
1342 **Figure 43.** Time series of AERONET (black lines) and simulated daily AOD (at wavelength of 675

1343 nm) during 2013 – 2017. Normalized mean bias (NMB) and correlation ( $r$ ) statistics between the



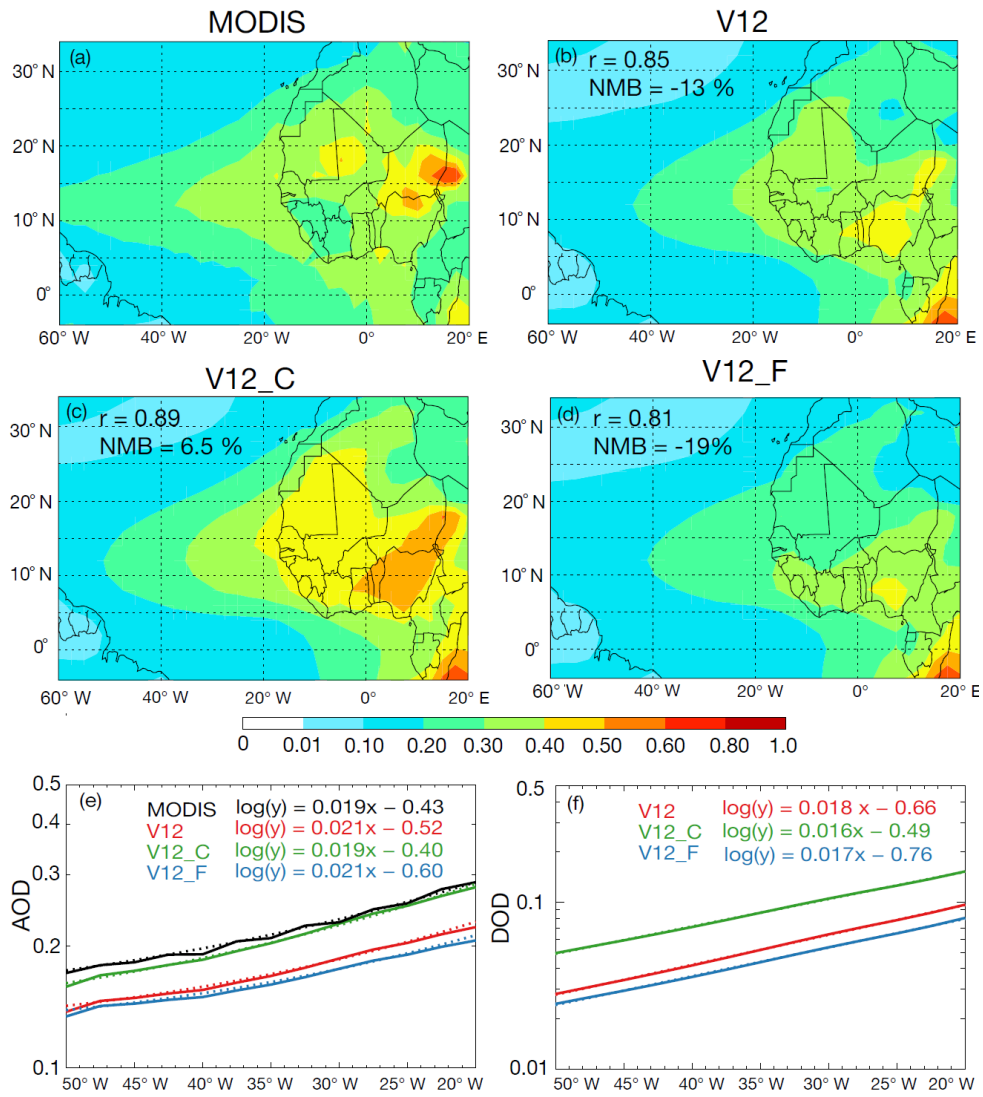
1344 AERONET and simulated data are shown as inset.

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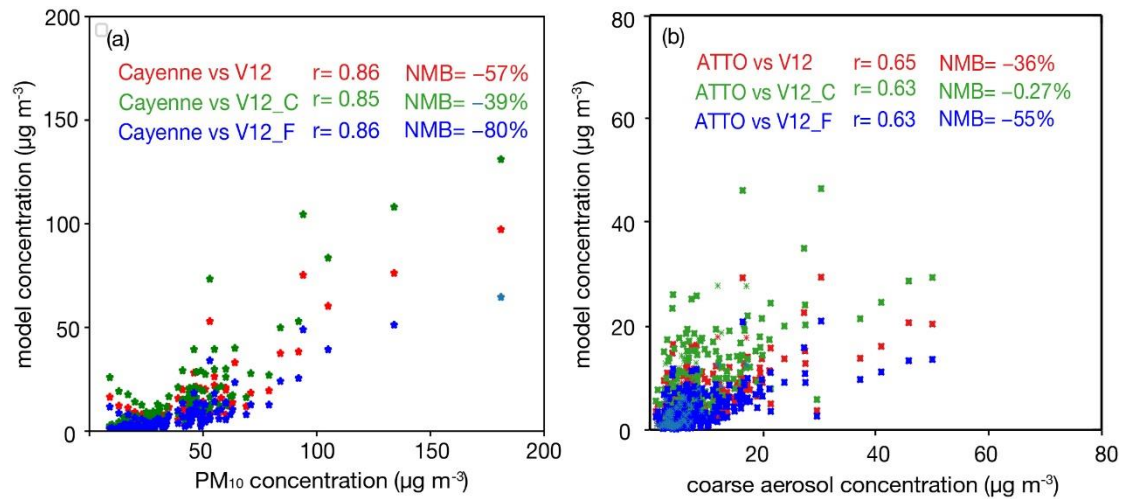
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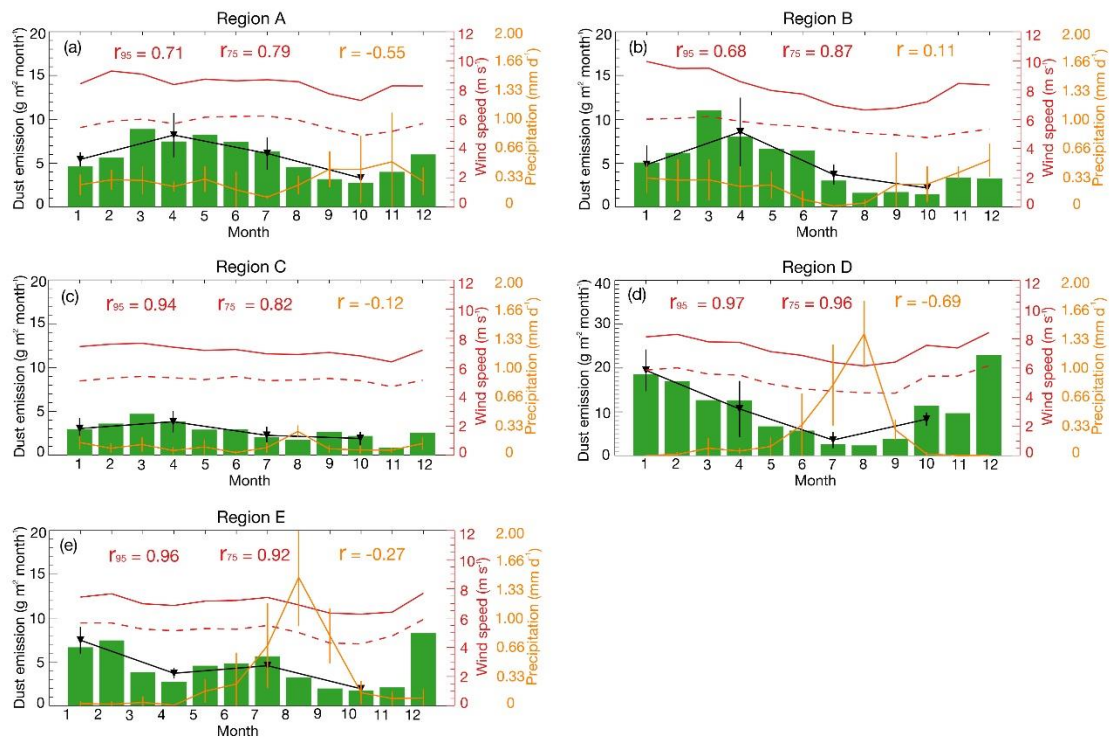
1349 **Figure 54.** (a) – (d) Spatial distributions of observed and simulated AOD (at 550 nm) over the  
 1350 region of 60° W – 20° E and 10° N – 35° N averaged over 2013 – 2017. Normalized mean bias  
 1351 (NMB) and correlation coefficient (r) between the simulations and MODIS AOD are shown as inset.  
 1352 (e) MODIS (black) and simulated (color) AOD and (f) simulated dust optical depth (DOD) at 550  
 1353 nm along the transect from 20° to 50° W, averaged over 5° S – 25° N for the period 2013 – 2017.  
 1354 The solid lines represent averaged data and the dashed lines are the logarithmic trend lines.

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**Figure 65.** Scatter plots of (a) observed  $\text{PM}_{10}$  and simulated dust concentrations at Cayenne site during wet season of 2014 and (b) observed coarse aerosol ( $\text{PM}_{1-10}$ ) and simulated dust concentrations at ATTO site during wet season of 2014-2016. Normalized mean bias (NMB) and correlation ( $r$ ) statistics between the observation and simulation are shown as inset.

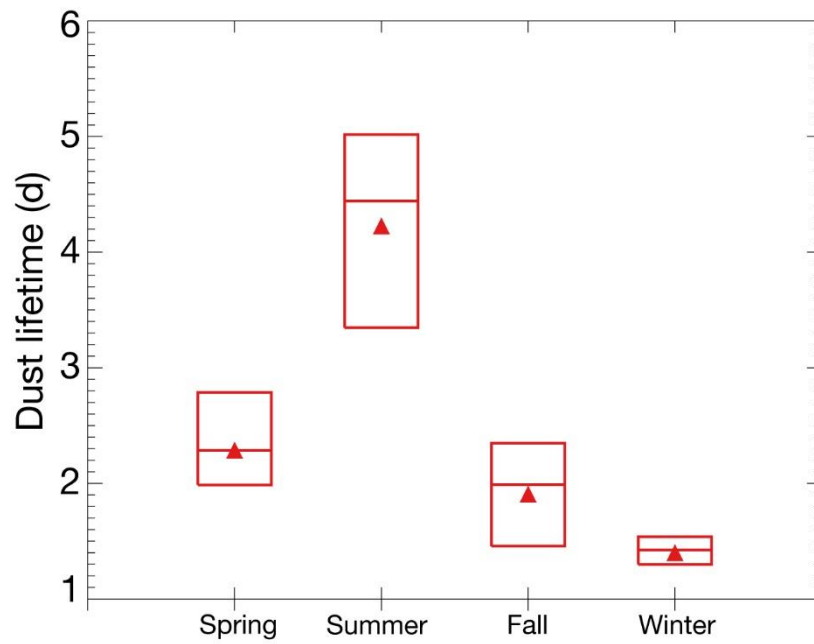


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**Figure 6.** Monthly dust emission fluxes together with the 95<sup>th</sup> percentile hourly 10-m wind speeds (red solid lines), the 75<sup>th</sup> percentile hourly 10-m wind speeds (red dotted lines) and precipitation (yellow lines) over the five major source regions averaged from 2013 to 2017. Seasonal emission fluxes of dust are also shown as black lines. The correlation coefficients ( $r$ ) between the dust emission fluxes and different meteorological variables are also shown in each panel.

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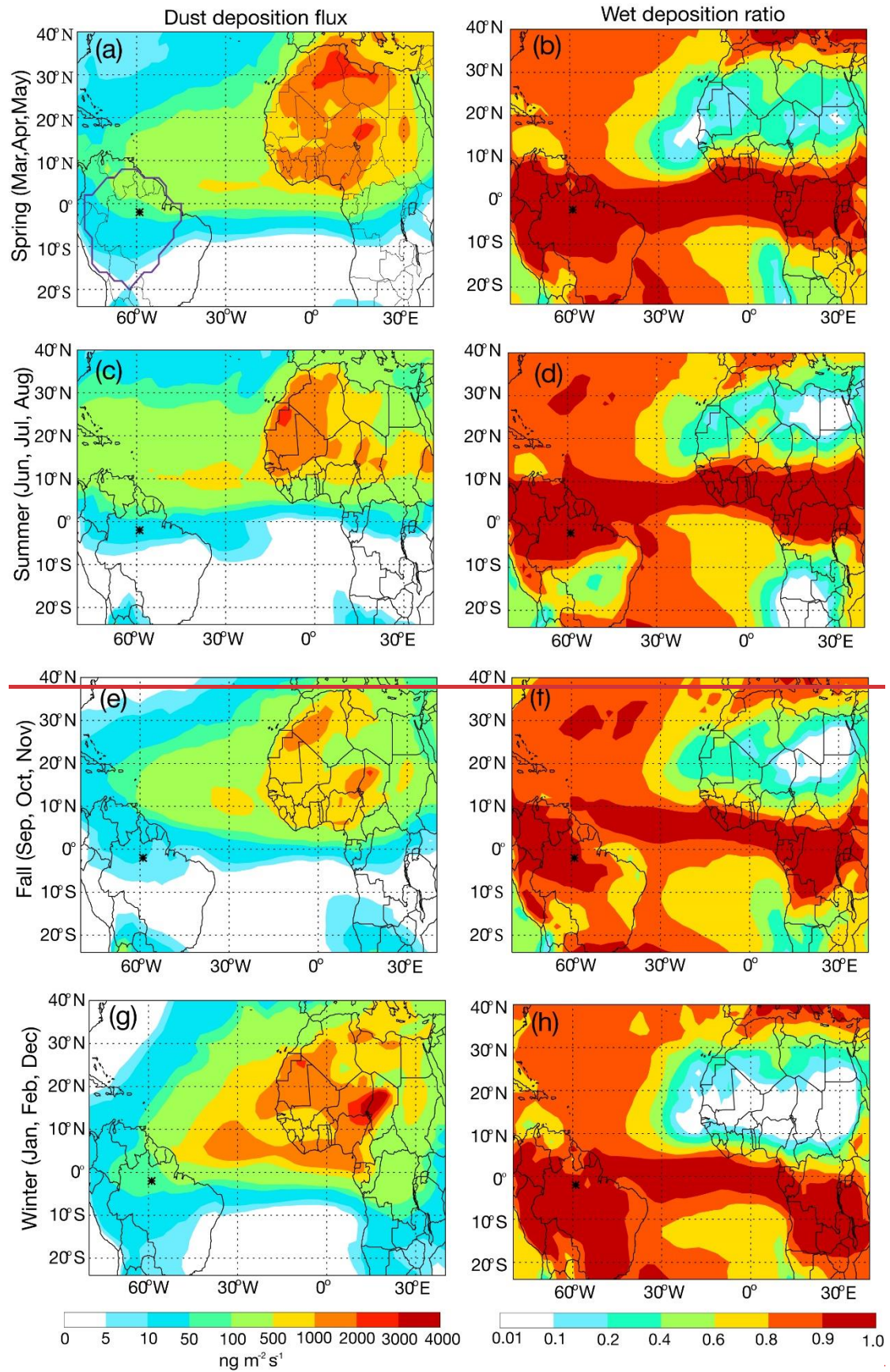
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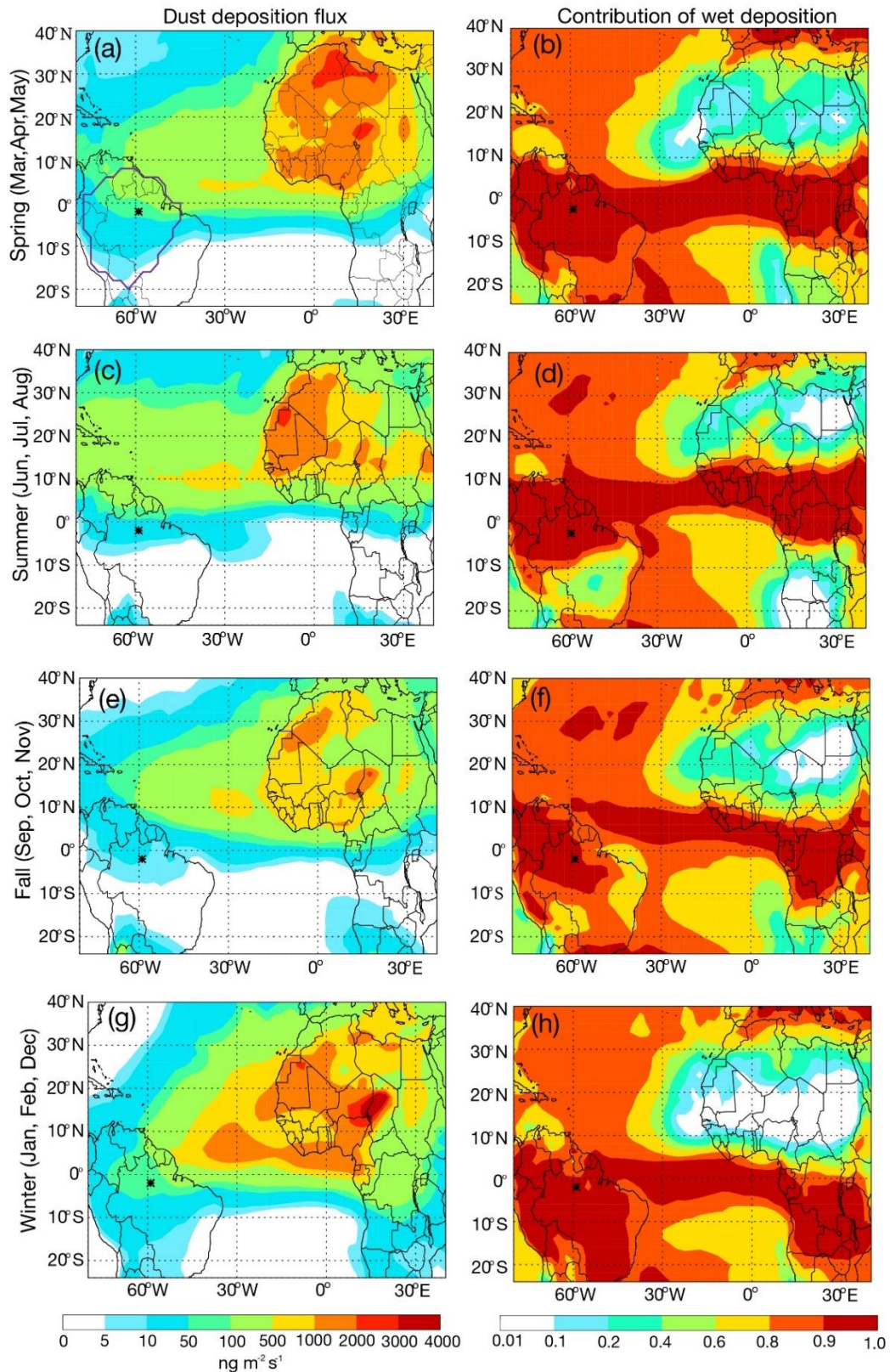
**Figure 7.** Seasonal e-folding lifetime derived from the logarithm of simulated dust column burden against travel time along the transect from 20° W to 50° W averaged over 5° S – 25° N during the period of 2013 – 2017. The triangles represent the mean values, and the bottom and top sides of the boxes represent the minimums and maximums.





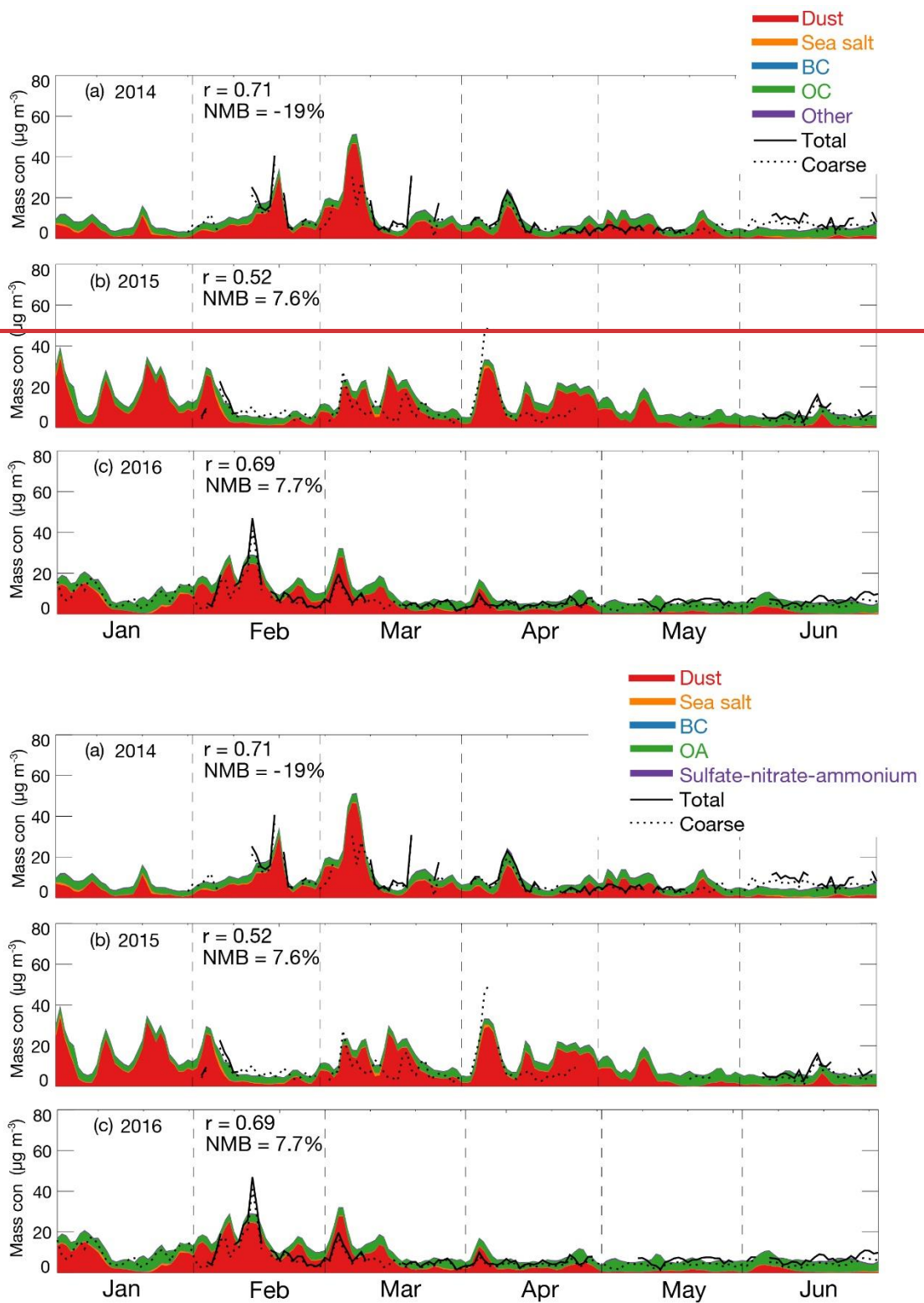
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1378 **Figure 8.** Simulated seasonal (left) dust deposition fluxes and (right) contribution of wet deposition  
 1379 during 2013-2017. The ATTO site is marked as asterisk. The region of the Amazon Basin is defined  
 1380 by purple lines in [Fig. 7a](#) [Figure 8a](#).



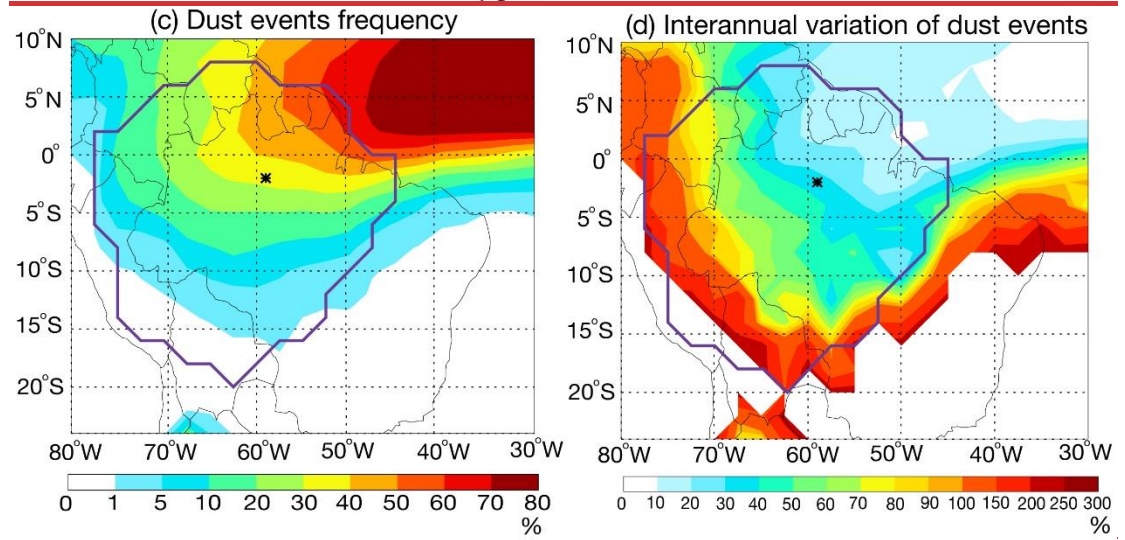
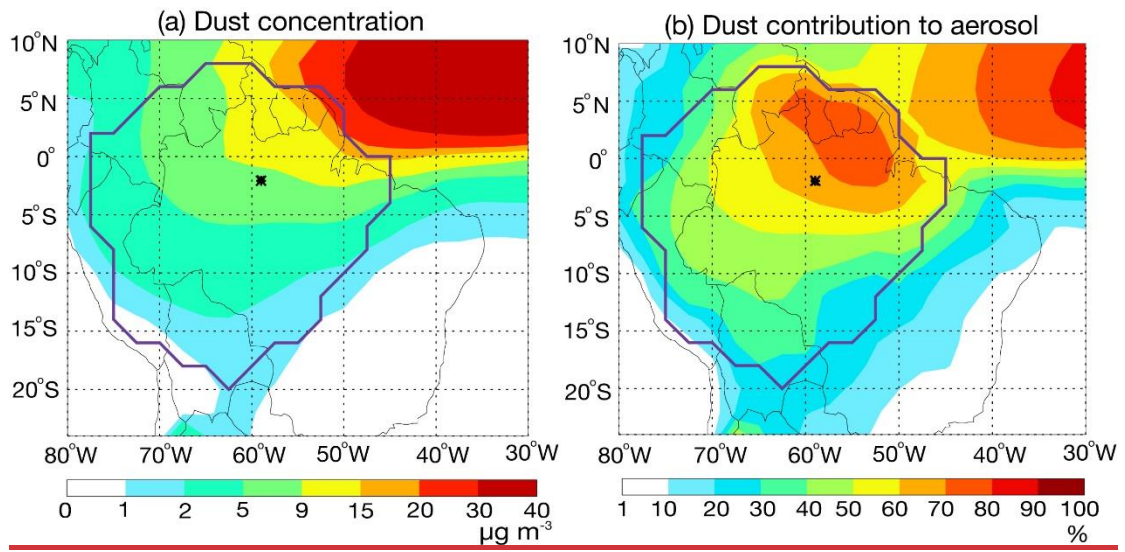
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1383 **Figure 9.** Time series of observed coarse and total aerosol mass concentrations and simulated  
 1384 aerosol species concentrations at the ATTO site from January to June in (a) 2014, (b) 2015, and (c)  
 1385 2016. Model results are separated into different species shown as stacked areas. Normalized mean  
 1386 bias (NMB) and correlation coefficient ( $r$ ) between the observed coarse aerosols and simulated dust  
 1387 concentrations are shown as inset.

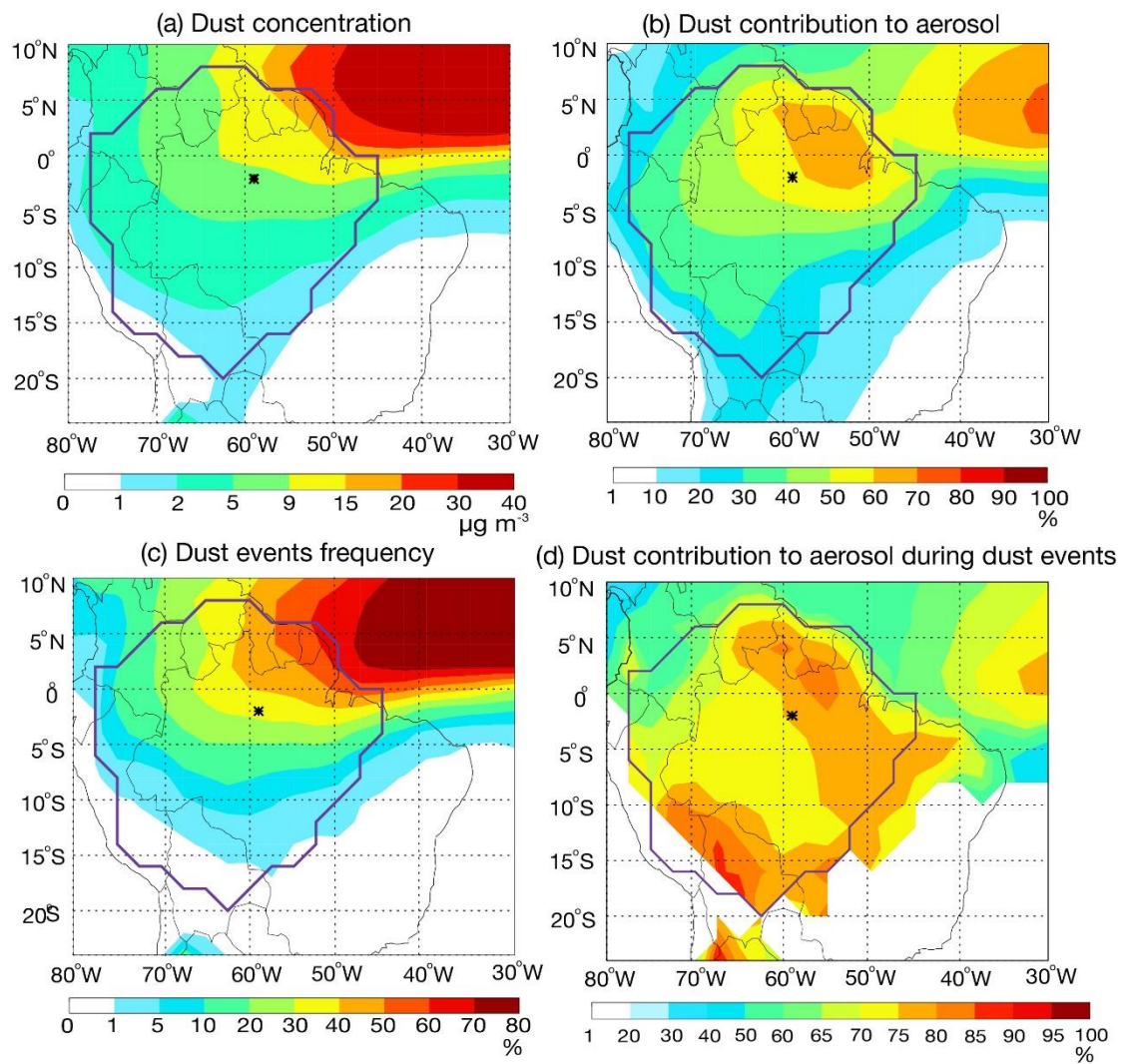


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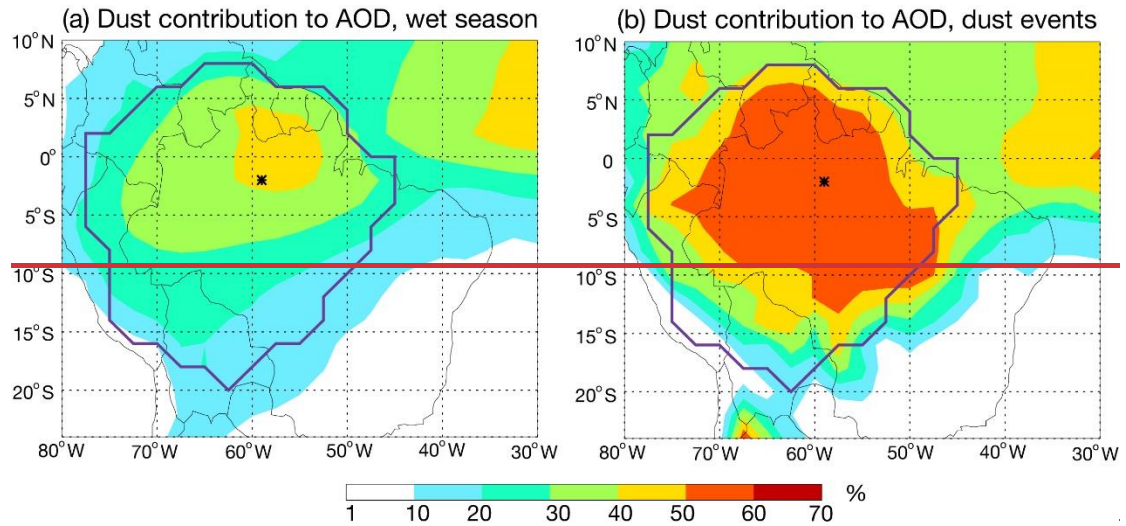
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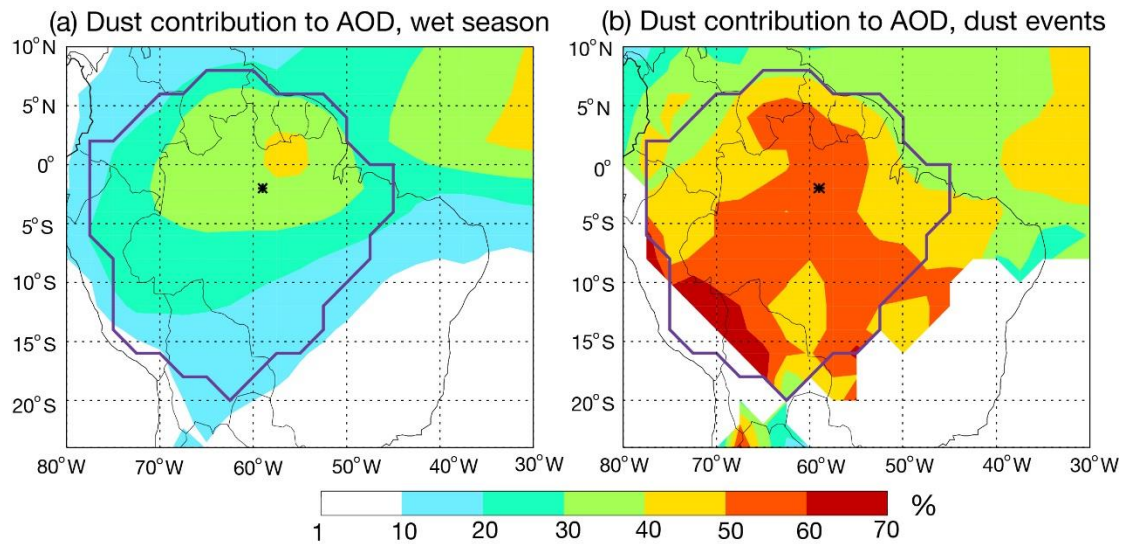
**Figure 10.** Dust impact over the Amazon Basin in the wet season of 2013 – 2017: (a) simulated surface dust ~~concentration and concentrations,~~ (b) ~~its dust~~ contribution to surface aerosol ~~concentration over the Amazon Basin in the wet season of 2013–2017.concentrations,~~ (c) the frequency of dust events, and (d) ~~its interannual variation (namely relative standard deviation)~~ dust contribution to surface aerosol concentrations during ~~the same period~~ dust events. The location of ATTO site is marked as asterisks. The region of Amazon Basin is marked by purple lines.



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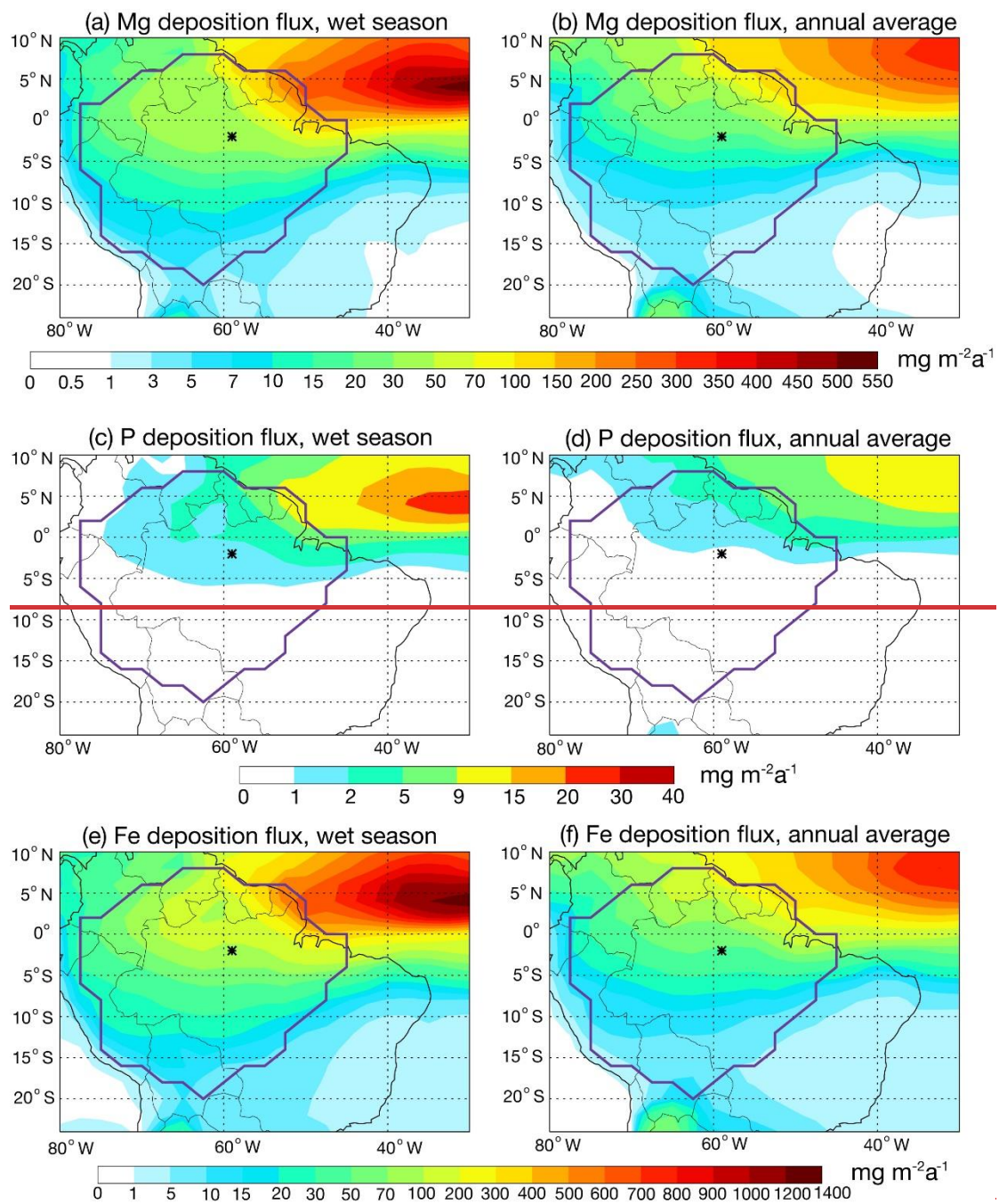
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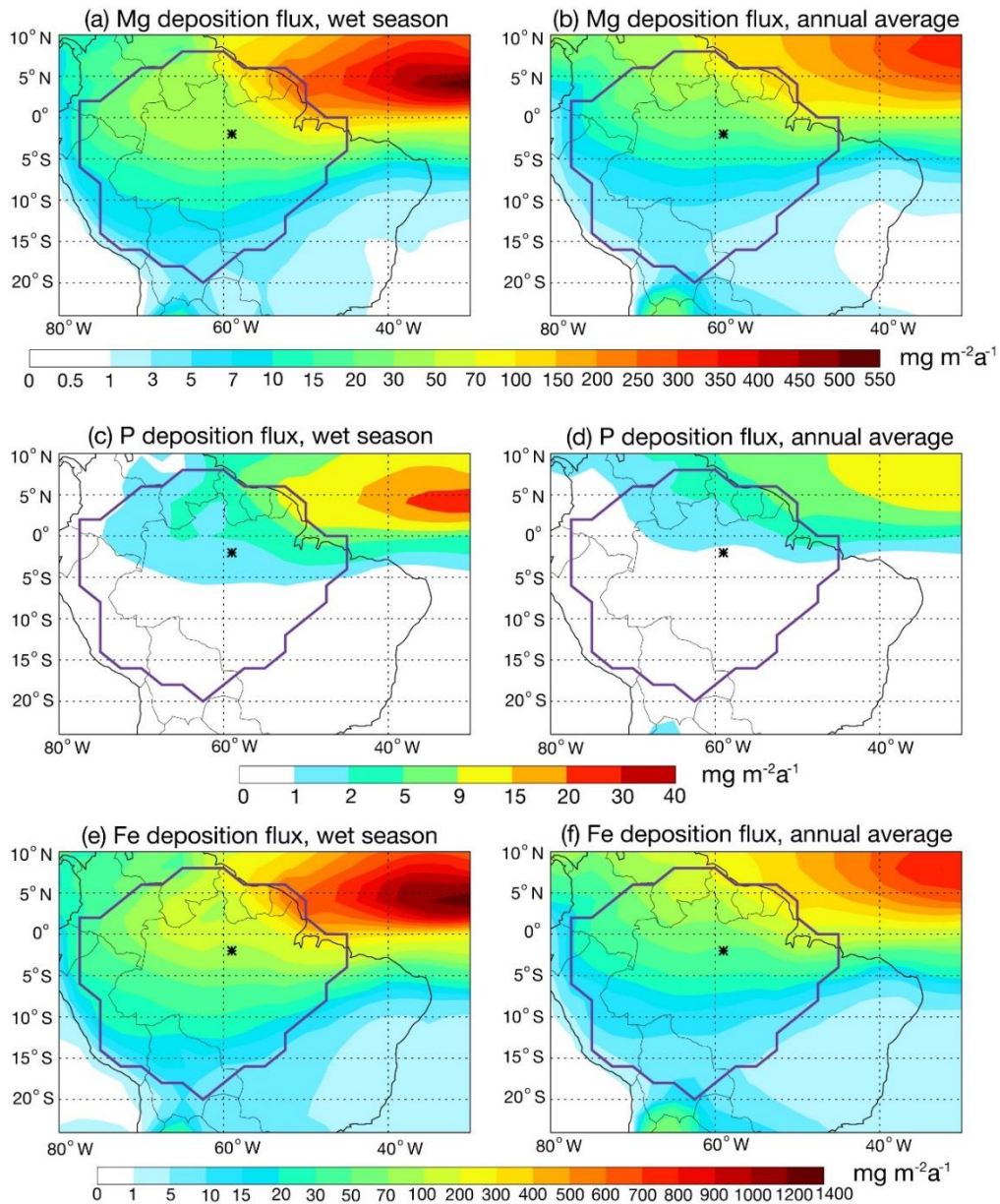
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**Figure 11.** Dust contribution to total AOD at 675.550 nm over the Amazon Basin averaged over the (a) wet season and (b) dust events during 2013--2017. The region of Amazon Basin is marked by purple lines.--







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1413 **Figure 12.** Magnesium deposition flux in (a) wet season and (b) annual averaged from 2013 to 2017.

1414 Phosphorus deposition flux in (c) wet season and (d) annual averaged from 2013 to 2017.

1415 Iron deposition flux in (e) wet season and (f) annual averaged from 2013 to 2017. The location of ATTO

1416 site is marked as asterisks. The region of Amazon Basin is marked by purple lines.

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