

1 **Improved model of isoprene emissions in Africa using OMI satellite observations of**  
2 **formaldehyde: implications for oxidants and particulate matter**

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22 **Abstract**

23 We use a 2005-2009 record of isoprene emissions over Africa derived from OMI satellite  
24 observations of formaldehyde (HCHO) to better understand the factors controlling isoprene  
25 emission in the continent and evaluate the impact on atmospheric composition. OMI-derived  
26 isoprene emissions show large seasonality over savannas driven by temperature and leaf area  
27 index (LAI), and much weaker seasonality over equatorial forests driven by temperature. The  
28 commonly used MEGAN (version 2.1) global isoprene emission model reproduces this  
29 seasonality but is biased high, particularly for equatorial forests, when compared to OMI and  
30 relaxed-eddy accumulation measurements. Isoprene emissions in MEGAN are computed as the  
31 product of an emission factor  $E_o$ , LAI, and activity factors dependent on environmental variables.  
32 We use the OMI-derived emissions to provide improved estimates of  $E_o$  that are in good  
33 agreement with direct leaf measurements from field campaigns ( $r = 0.55$ , bias = -19%). The  
34 largest downward corrections to MEGAN  $E_o$  values are for equatorial forests and semi-arid  
35 environments, and this is consistent with latitudinal transects of isoprene over West Africa from  
36 the AMMA aircraft campaign. Total emission of isoprene in Africa is estimated to be 77 Tg C a<sup>-1</sup>,  
37 compared to 104 Tg C a<sup>-1</sup> in MEGAN. Simulations with the GEOS-Chem oxidant-aerosol  
38 model suggest that isoprene emissions increase mean surface ozone in West Africa by up to 8  
39 ppbv, and particulate matter by up to 1.5  $\mu\text{g m}^{-3}$ , due to coupling with anthropogenic influences.  
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## 42 **1. Introduction**

43 Isoprene is the dominant biogenic non-methane volatile organic compound (NMVOC)  
44 emitted by vegetation, accounting for about 50% of global NMVOC emissions in current  
45 inventories (Olivier et al., 1996; Guenther et al., 2006). Isoprene affects the oxidative capacity of  
46 the atmosphere through reaction with OH (Ren et al., 2008; Lelieveld et al., 2008) and as a  
47 precursor of O<sub>3</sub> (Trainer et al., 1987). It is also an important precursor for secondary organic  
48 aerosols (SOA) (Claeys et al., 2004) and a temporary reservoir for nitrogen oxide radicals (NO<sub>x</sub>  
49 ≡ NO + NO<sub>2</sub>) by formation of organic nitrates (Paulot et al., 2012). Isoprene thus has a range of  
50 impacts on air quality and climate that need to be included in atmospheric composition models.  
51 The widely used MEGAN global emission model (Guenther et al., 2006, 2012) indicates that  
52 80% of global isoprene emission takes place in the tropics and 25% in Africa, but there are large  
53 uncertainties in these estimates due to lack of data. In previous work we developed a method to  
54 estimate isoprene emissions from Africa on the basis of observations of formaldehyde (HCHO)  
55 from the OMI satellite instrument (Marais et al., 2012). Here we use our OMI-derived isoprene  
56 emissions evaluated with local data to better understand the factors controlling isoprene  
57 emissions in Africa, improve emission estimates for different African plant functional types  
58 (PFTs), and assess the implications for atmospheric oxidants and aerosols.

59 Isoprene produced in the chloroplasts of plants is released to the atmosphere via the  
60 stomata of leaves (Sharkey and Yeh, 2001). Above-canopy emission fluxes  $E_{\text{ISOP}}$  depend on  
61 plant species, foliage density, leaf age, temperature, photosynthetically active radiation (PAR),  
62 and water stress (Guenther et al., 1995). This is commonly represented in isoprene emission  
63 models by multiplying an emission factor  $E_o$  defined for each PFT at standard conditions with an

64 ensemble of coefficients describing the sensitivity to local environmental variables. In the  
65 MEGAN (version 2.1) inventory (Guenther et al., 2012) this is given as

$$66 \quad E_{\text{ISOP}} = E_o \times C_{\text{CE}} \times \text{LAI} \times \gamma_{\text{PAR}} \times \gamma_T \times \gamma_{\text{AGE}} \times \gamma_{\text{SM}} \quad (1).$$

67 where LAI is the leaf area index ( $\text{m}^2$  leaf surface per  $\text{m}^2$  of Earth surface) and the dimensionless  
68 activity factors  $\gamma$  describe the sensitivity to above-canopy radiation (PAR), air temperature ( $T$ ),  
69 leaf age distribution (AGE), and soil moisture (SM). The coefficient  $C_{\text{CE}} = 1.3$  ( $\text{m}^2$  Earth surface  
70 per  $\text{m}^2$  leaf surface) enforces  $E_{\text{ISOP}} = E_o$  under standard conditions, which for MEGAN are  
71 defined as  $T = 303$  K,  $\text{PAR} = 1000$   $\mu\text{mol photons m}^{-2} \text{s}^{-1}$ , a canopy with  $\text{LAI} = 5$   $\text{m}^2 \text{m}^{-2}$ , and leaf  
72 age distribution of 80% mature, 10% growing, and 10% senescing leaves, and volumetric soil  
73 moisture of  $0.3$   $\text{m}^3 \text{m}^{-3}$ .

74 Isoprene emission data for African vegetation are very limited, and emission models  
75 require extrapolation of data from other continents and across plant species (Guenther et al.,  
76 2006; 2012). This can lead to substantial errors, as differences in isoprene fluxes within and  
77 across plant species are large. Uncertainty in the distribution of land cover (PFT) adds to the  
78 uncertainty (Pfister et al., 2008).

79 Space-based observations of HCHO, a high-yield oxidation product of isoprene, have  
80 been used in a number of studies to infer isoprene emissions and evaluate inventories globally  
81 (Shim et al., 2005; Stavrou et al., 2009a; 2009b) and regionally in Southeast Asia (Fu et al.,  
82 2007), South America (Barkley et al., 2008), North America (Palmer et al., 2003; 2006; Millet et  
83 al., 2008), Europe (Dufour et al., 2009; Curci et al., 2010), and Africa (Marais et al., 2012).  
84 These studies have confirmed temperature as the dominant factor controlling month-to-month  
85 variability of isoprene emissions across North America (Palmer et al., 2006; Millet et al., 2008)  
86 and Amazonia (Barkley et al., 2008). Leaf phenology and PAR were found to be additional

87 important drivers of isoprene emission seasonality in Amazonia (Barkley et al., 2008; 2009).  
88 Stavrakou et al. (2009a) found that water stress reduces isoprene emissions in southern Africa  
89 during the dry season. Here we use our previous work for Africa (Marais et al., 2012) to better  
90 understand the factors controlling isoprene emissions across the African continent and evaluate  
91 and improve the MEGANv2.1 emission inventory.

92

## 93 **2. OMI-derived isoprene emissions in Africa**

94 The derivation of isoprene emissions in Africa using OMI HCHO data is described in  
95 Marais et al. (2012) and summarized briefly here. OMI is a UV/VIS solar backscatter instrument  
96 on the Aura polar sun-synchronous satellite launched in 2004 (Levelt et al., 2006). It has a 13×24  
97 km<sup>2</sup> nadir pixel resolution, daily global coverage through cross-track viewing, and 13:30 local  
98 time (LT) overpass. HCHO slant columns are obtained from Version 2.0 (Collection 3) retrievals  
99 for 2005–2009 ([http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omhcho\\_v003.shtml](http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omhcho_v003.shtml)).  
100 They are corrected for instrument drift and converted to vertical columns using local air mass  
101 factors (AMF) for the scattering atmosphere (Palmer et al., 2001) with vertical HCHO profiles  
102 from the GEOS-Chem chemical transport model (CTM) v9-01-03 (<http://www.geos-chem.org>)  
103 and scattering weights from the LIDORT radiative transfer model (Spurr et al., 2001).

104 HCHO enhancements over Africa primarily originate from isoprene emission, biomass  
105 burning, and fuel combustion. Scenes affected by biomass burning are excluded on the basis of  
106 MODIS satellite observations of fire counts and OMI satellite observations of aerosol absorption  
107 optical depth (AAOD) (Torres et al., 2007). Scenes affected by gas flaring are excluded on the  
108 basis of a specialized hotspot product from the AATSR satellite sensor (Casadio et al., 2012) and

109 this leads to the exclusion of much of Nigeria where that source is particularly large and urban  
110 and industrial sources may contribute as well (Marais et al., 2014).

111 Marais et al. (2012) thus obtained a 2005-2009 monthly data set of vertical HCHO  
112 columns with  $1 \times 1^\circ$  spatial resolution screened against biomass burning and anthropogenic  
113 influences and thus attributable to isoprene emissions (Fig. 1, left panel). They used GEOS-  
114 Chem to derive the sensitivity,  $S$ , of the HCHO column ( $\Omega_{\text{HCHO}}$ ) at 12-15 LT to a perturbation  $\Delta$   
115 in isoprene emission ( $S = \Delta\Omega_{\text{HCHO}} / \Delta E_{\text{ISOP}}$ ). Values of  $S$  increase linearly with  $\text{NO}_x$  under low-  
116  $\text{NO}_x$  conditions (boundary-layer  $\text{NO}_x < 500$  pptv) and remain constant above 500 pptv  $\text{NO}_x$ . The  
117 local regime for individual scenes was determined from concurrent observations of OMI  
118 tropospheric  $\text{NO}_2$  columns. Scenes affected by smearing (displacement of HCHO from the  
119 isoprene emission source) were diagnosed with anomalously high values of  $S$  and excluded from  
120 the data set. See Marais et al. (2012) for further details.

121 Marais et al. (2012) obtained in this manner a monthly isoprene emission inventory for  
122 2005-2009 on a  $1 \times 1^\circ$  grid (Fig. 1, center panel). The OMI overpass is at 13:30 LT and the  
123 corresponding isoprene emissions are for 12-15 LT, typically the diurnal maximum. Also shown  
124 in Fig. 1 is the MODIS IGBP land cover map (Friedl et al., 2002). Dominant vegetation types in  
125 Africa are roughly defined by latitudinal bands, with evergreen (broadleaf) trees along the  
126 Equator successively transitioning to the north and south to woody savannas (30-60% tree  
127 coverage), savannas (10-30%), grasslands, and deserts. The HCHO column data follow this  
128 vegetation gradient and so do the inferred isoprene emissions.

129 Marais et al. (2012) presented a detailed error characterization of their OMI-derived  
130 isoprene emissions. Spectral fitting of the HCHO column has an error standard deviation of  $8 \times$   
131  $10^{15}$  molecules  $\text{cm}^{-2}$  for individual observations. Relating the fitted slant HCHO columns to

132 isoprene emissions incurs errors in the AMF estimate (20%), the isoprene oxidation mechanism  
133 (15%), the use of OMI NO<sub>2</sub> to obtain *S* under low-NO<sub>x</sub> conditions (20-40%), and smearing (30%  
134 for high-NO<sub>x</sub> conditions, 30-70% for low-NO<sub>x</sub>). The resulting error in isoprene emission  
135 estimates for individual scenes, adding in quadrature all error contributions, is 40% for high-NO<sub>x</sub>  
136 conditions and 40-90% for low-NO<sub>x</sub> (Marais et al., 2012). A monthly mean estimate for a 1×1°  
137 gridsquare typically averages 3000 individual scenes. Averaging reduces the error though only to  
138 the extent that the error components are random.

139

### 140 **3. Evaluation with canopy flux measurements**

141 Canopy-scale isoprene flux measurements by relaxed-eddy accumulation (REA) are  
142 available from a few African field campaigns (Table 1a). Figure 2 compares OMI-derived  
143 isoprene emissions to REA measurements over equatorial evergreen trees (Greenberg et al.,  
144 1999; Serça et al., 2001), woody savannas (Greenberg et al., 1999), and savannas (Harley et al.,  
145 2003) in central and southern Africa (sites 1-4 in Fig. 1 and Table 1a). Also shown are the values  
146 calculated using Eq. (1) with MEGAN v2.1 emission factors, combined Terra and Aqua MODIS  
147 LAI (Yang et al., 2006), and the Goddard Earth Observing System (GEOS-5) assimilated  
148 meteorological data. The Serça et al. (2001) and Harley et al. (2003) measurements (sites 1 and  
149 4) are from walk-up towers with a flux footprint of about 600 m, while the Greenberg et al.  
150 (1999) measurements (sites 2 and 3) are from aircraft with a flux footprint of ~100×100 km<sup>2</sup> at  
151 site 2 and 30×30 km<sup>2</sup> at site 3. All values in Fig. 2 are for 12-15 LT. REA fluxes at sites 2-3,  
152 obtained in the morning (9:30-11:30), are increased by a factor of 1.4 as a diurnal correction for  
153 temperature and PAR following MEGAN.

154 No correction is applied to the REA flux measurements to account for interannual  
155 variability between 1996 (sites 1-3), 2001 (site 4), and the satellite observation period (2005-  
156 2009). As will be discussed in Sect. 4, temperature is the dominant modulator of isoprene  
157 emissions in Africa and it does not drive significant interannual variability except in northern  
158 savannas in August (not represented in Fig. 2).

159 OMI and MEGAN are sampled for the  $1 \times 1^\circ$  gridsquare coincident with the observation  
160 site and for the corresponding months. Interannual variability is of similar magnitude in the  
161 OMI-derived and MEGAN data at sites 1 (November), 2 and, 3 where multi-year OMI data are  
162 available. The variability is driven in MEGAN predominantly by temperature. At site 3 there are  
163 no OMI data in the months of observation (November-December) because of biomass burning  
164 interference and we show instead OMI-derived emissions in September-October, which should  
165 be similar to November-December at this site according to MEGAN.

166 For the equatorial evergreen tree sites in central Africa (sites 1 and 2) OMI-derived  
167 isoprene emissions are on average 2 times higher than the REA measurements and MEGAN is 5  
168 times higher. The flux tower sampled vegetation with a relatively low fraction of isoprene  
169 emitters. Nearby landscapes include monodominant stands of the *Gilbertiodendron* trees that  
170 have a high isoprene emission factor (Serça et al., 2001). The distribution of this tree species  
171 beyond the sampling domain is uncertain and application of its emission factor to land cover in  
172 equatorial Africa contributes to the overestimate in MEGAN. OMI and MEGAN reproduce the  
173 March-November decline at site 1 and this is driven in MEGAN by temperature.

174 Fluxes at site 2, where the REA sampling footprint is similar to OMI, have large spatial  
175 variability implying that differences in the sampling footprint contribute to discrepancies at other  
176 sites. Greenberg et al. (1999) applied a positive correction of ~20% to flux measurements at sites

177 2-3 to account for the transport of isoprene that was not accumulated in the two REA reservoirs.  
178 A similar negative bias may affect measurements at sites 1 and 4 but the reported values have not  
179 been adjusted. The aircraft REA measurements (sites 2 and 3) may have an additional negative  
180 bias of ~25% due to the vertical flux divergence between the measurement altitude and the  
181 surface (Karl et al., 2013).

182 At the woody savanna site OMI is 2.2 times higher than the REA measurement (1.8 times  
183 higher if a 25% upward correction is applied to the REA measurement), while MEGAN is 8  
184 times higher. At the savanna site OMI is 1.3 times higher than the REA flux measurement while  
185 MEGAN is 2.4 times higher. The discrepancy at site 4 is partly due to the low (<10%) proportion  
186 of isoprene emitting vegetation within the flux tower footprint as compared to ~35% for  
187 savannas surveyed at surrounding field sites (Harley et al., 2003).

188 Overall the REA flux measurements indicate canopy-scale isoprene emissions that are  
189 somewhat lower than derived from OMI and much lower than derived from MEGAN. As we  
190 will show in Sect. 5, landscape-level isoprene emission factors measured during African field  
191 campaigns are more consistent with OMI.

192

#### 193 **4. Seasonality of isoprene emissions in Africa**

194 We examined the factors driving the seasonality of OMI-derived isoprene emissions,  
195 focusing on three seasonally and ecologically coherent regions in Africa where emissions are  
196 highest (Fig. 3): (1) equatorial forests dominated by tropical broadleaf evergreen trees, (2)  
197 northern savannas (including woody), and (3) southern savannas (including woody). Figure 4  
198 shows the seasonality of OMI-derived isoprene emissions for these three regions together with  
199 MODIS LAI and GEOS-5 2-m air temperature. Isoprene emissions and 2-m temperature are for

200 12-15 LT and MODIS LAI is the combined Terra and Aqua product (Yang et al., 2006). Multi-  
201 year averages (2005-2009) are shown; the regionally averaged interannual variability is small,  
202 except over northern savannas in August as discussed below. Soil moisture and PAR are not  
203 included in Fig. 4 as soil moisture only appears to affect southern savannas during the dry  
204 season, and PAR in southern and northern savannas is convolved with temperature.

205 OMI-derived isoprene emissions for equatorial forests are a factor of 2 lower than  
206 MEGAN and both show similar weak seasonality, with a decline from March to November that  
207 is consistent with Serça et al. (2001) (Fig. 2) and is driven in MEGAN principally by  
208 temperature. Although LAI exhibits similar seasonality, it remains above  $3.5 \text{ m}^2 \text{ m}^{-2}$  year-round  
209 and MEGAN is not sensitive to LAI values above  $2\text{-}3 \text{ m}^2 \text{ m}^{-2}$  due to shading of lower-canopy  
210 leaves (Guenther et al., 2006).

211 Availability of OMI-derived isoprene emission data for the northern savannas is limited  
212 to April-November because of pervasive biomass burning influence during the December-March  
213 dry season. Emissions are maximum in April, at the beginning of the wet season, and minimum  
214 in August when the West African Monsoon (WAM) is fully developed over the continent  
215 (Janicot et al., 2008) resulting in cooler temperatures. OMI-derived emissions largely follow  
216 temperature over the April-November period. Year-to-year variability in the WAM affects  
217 temperature in August, leading to interannual variability in August OMI isoprene emissions over  
218 the 2005-2009 period that is correlated with temperature ( $r = 0.55$ ).

219 The complete seasonality simulated by MEGAN in northern savannas shows low  
220 isoprene emissions in the December-March dry season when LAI is less than  $1 \text{ m}^2 \text{ m}^{-2}$ , and a  
221 broad maximum in the April-November wet season as the August minimum in temperature is  
222 compensated by a corresponding maximum in LAI. MODIS LAI in the northern savannas is less

223 than  $2.5 \text{ m}^2 \text{ m}^{-2}$  year-round, sufficiently low that the MEGAN dependence on LAI does not  
224 saturate (Guenther et al., 2012). However, MODIS may underestimate LAI in West Africa  
225 during the wet season because of cloud contamination (Gessner et al., 2013).

226 OMI-derived emissions for southern savannas are in close agreement with MEGAN,  
227 featuring a minimum in the winter dry season and a maximum in the summer wet season. The  
228 seasonal minimum follows that of temperature (June-July) with a 1-month lag that reflects the  
229 very dry conditions in July-September. We find that LAI and temperature are both important for  
230 driving the seasonality of isoprene emissions in southern savannas.

231

## 232 **5. Satellite-derived isoprene emission factors for Africa**

233 The general ability of MEGAN to reproduce the seasonal variation of OMI-derived  
234 isoprene emissions suggests that the MEGAN activity factors ( $\gamma$  in Eq. (1)) are appropriate for  
235 conditions in Africa, with temperature and LAI as the principal drivers. The large MEGAN bias  
236 over equatorial forests and northern savannas can therefore be attributed to an overestimate of  
237 emission factors ( $E_o$  in Eq. (1)).

238 The emission factors in MEGAN represent isoprene fluxes for a canopy with leaves at  
239 standard conditions of air temperature ( $T = 303 \text{ K}$ ) and light ( $\text{PAR} = 1000 \mu\text{mol photons m}^{-2} \text{ s}^{-1}$ ).  
240 They are gridded using a detailed regional land cover map for Africa south of the Equator (Otter  
241 et al., 2003) and the Olson et al. (2001) global ecoregion data in the north (Guenther et al., 2006).  
242 Here we infer emission factors ( $E_0$ ) from the OMI-derived canopy-level isoprene emission data  
243 ( $E_{\text{ISOP}}$ ) using Eq. (1). In so doing we only consider data with individual activity factors  $\gamma$  in the  
244 range 0.5-1.5 so as to avoid errors driven by large departures from standard conditions.

245 Figure 5 shows the resulting distributions of MEGAN and OMI emission factors  $E_o$  over  
246 Africa together with observations from the field campaigns of Table 1b. The latter are at the  
247 landscape-level and were obtained by scaling measured leaf-level isoprene fluxes for  
248 representative plant species with foliage density and species distribution data. Leaf-level  
249 measurements at standard conditions were obtained by using enclosure measurements with  
250 controlled temperature and PAR (Serça et al., 2001; Otter et al., 2002), or adjusting to standard  
251 conditions with MEGAN activity factors for temperature and PAR (Guenther et al., 1996). For  
252 the former an upward correction applied to the Serça et al. (2001) landscape emission factor  
253 accounts for isoprene fluxes obtained from shade-adapted leaves that have lower emissions at  
254 standard conditions than sunlit leaves (Guenther et al., 1999). Leaf-level fluxes of Klinger et al.  
255 (1998) were determined to be at standard conditions with coincident measurements of  
256 temperature and PAR, but we exclude the data from shaded leaves at dense forest sites.

257 We find from Fig. 5 remarkable agreement between OMI-derived emission factors and  
258 the field data ( $r = 0.55$ , OMI normalized mean bias = -19%). Woody savannas in Zambia and  
259 savannas in South Africa have large variability in  $E_o$  (0.5-4.5 mg C m<sup>-2</sup> h<sup>-1</sup>) that is reproduced by  
260 OMI. The two sites in Botswana have low emission factors as the site to the north is dominated  
261 by monoterpene emitting mopane vegetation, while the site to the south is predominantly  
262 shrubland (Otter et al., 2002).

263 Differences between OMI and MEGAN emission factors are largest for equatorial  
264 forests, and the field enclosure observations are in good agreement with OMI and much lower  
265 than MEGAN. The equatorial forest enclosure measurements are used in MEGAN to estimate  
266 emission factors there, but a large positive correction is applied to account for leaf enclosure  
267 measurements of shade-adapted leaves. Our OMI-derived emission factors do not support such a

268 correction, and this is also reflected in the MEGAN overestimate of REA flux measurements  
269 (Fig. 2). OMI emission factors for equatorial forests are larger in the west than east and this may  
270 result from differences in the proportion of isoprene emitting species. The west is dominated by  
271 dry tropical forests, while the east is dominated by permanently or seasonally flooded forests  
272 (White, 1983).

273 Table 2 shows mean isoprene emission factors for individual PFTs as obtained by  
274 mapping the data from Fig. 5 onto the MODIS IGBP land map (Fig. 1; Friedl et al., 2002) and  
275 the Global Land Cover (GLC) 2000 land map (Mayaux et al., 2006). The distribution of MODIS  
276 IGBP woody savannas is spatially consistent with GLC 2000 broadleaf trees and MODIS IGBP  
277 savannas correspond to GLC 2000 shrubs interspersed with broadleaf trees and cultivated land.  
278 The GLC 2000 classification scheme is more consistent with the PFTs of MEGANv2.1  
279 (Guenther et al., 2012). OMI gives higher emission factors for forested vegetation than for  
280 grasslands but the difference is not as large as MEGAN and more consistent with the field  
281 enclosure observations. The largest differences between OMI and MEGAN emission factors are  
282 for broadleaf evergreen trees and for semi-arid vegetation (shrubs and herbs).

283 The OMI-derived emission factors in Fig. 5 can be used to improve the MEGAN  
284 isoprene emission estimates as computed from Eq. (1). Figure 6 compares the resulting isoprene  
285 concentrations simulated by GEOS-Chem with a latitudinal profile of isoprene concentration  
286 measurements below 1 km across West Africa during the AMMA wet season aircraft campaign  
287 in July-August 2006 (Murphy et al., 2010). There is a strong vegetation gradient along the  
288 AMMA flight track from the Gulf of Guinea to Benin woodlands to arid conditions in the north  
289 that is reflected in the isoprene data. Simulated isoprene in GEOS-Chem is a factor of 2 too low  
290 over Benin woodlands likely due to a seasonal low bias in MODIS LAI over West Africa from

291 cloud contamination (Gessner et al., 2013). Isoprene emissions over the AMMA domain are not  
292 only sensitive to LAI, but also MEGAN emission factors (Ferreira et al., 2010). The OMI-  
293 derived emission factors are much better able to reproduce the latitudinal gradient than the  
294 original MEGAN emission factors, including in particular the decline to the north associated  
295 with increased aridity. Throughout Africa MEGAN emission factors are too high for semi-arid  
296 PFTs, as indicated by the overestimate in MEGAN for GLC 2000 sparse herb/shrub cover in  
297 Table 2.

298

## 299 **6. Implications for oxidants and aerosols**

300 We use the GEOS-Chem chemical transport model to (1) evaluate the change in  
301 atmospheric composition that results from replacing MEGAN emission factors with those  
302 obtained using OMI, and (2) determine the impact of isoprene emissions on aerosols and  
303 oxidants. GEOS-Chem includes the standard representation of oxidant-aerosol chemistry as  
304 described for example by Mao et al. (2010) with updates to the isoprene oxidation (Paulot et al.,  
305 2009a; 2009b).

306 Total annual isoprene emissions in Africa averaged over 2005-2009 using OMI-derived  
307 emission factors are  $77 \text{ Tg C a}^{-1}$ , as compared to  $104 \text{ Tg C a}^{-1}$  in MEGAN v2.1. The difference is  
308 mainly for the equatorial evergreen forest PFT in central Africa, where OMI-derived isoprene  
309 emissions are 2-3 times lower than MEGAN. GEOS-Chem using OMI-derived isoprene  
310 emissions indicates a factor of 4 increase in boundary layer OH concentrations over central  
311 Africa relative to MEGAN, a 4 ppbv increase in surface  $\text{O}_3$ , and 8 ppbv decrease in surface  
312 isoprene.

313 Figure 7 shows the effect of African isoprene emissions (using OMI-derived emission  
314 factors) on surface concentrations of daily maximum 8-hour average (MDA8) O<sub>3</sub>, particulate  
315 matter (PM), NO<sub>x</sub>, and OH. The largest effect on O<sub>3</sub> is over West Africa because of high  
316 anthropogenic, soil, and biomass burning NO<sub>x</sub> emissions (Marais et al., 2014). The largest effect  
317 on PM is also over West Africa and reflects the availability of high pre-existing primary PM  
318 from combustion (biomass burning and fuel) on which isoprene oxidation products can  
319 condense. NO<sub>x</sub> declines in West Africa and the tropics due to formation of isoprene nitrates.  
320 Loss of OH from reaction with isoprene is highest in the tropics where low levels of NO<sub>x</sub> limit  
321 the recycling of HO<sub>x</sub> radicals.

322

## 323 7. Conclusions

324 We used a 2005-2009 data set of monthly isoprene emissions in Africa derived from OMI  
325 satellite observations of formaldehyde (HCHO) to study the factors controlling these emissions  
326 in different areas of the continent. Our goal was to achieve a better representation of isoprene  
327 emission in chemical transport models (CTMs), in part through evaluation and improvement of  
328 the commonly used MEGAN emission inventory, and to examine the implications for oxidants  
329 and aerosols over the continent.

330 We began by evaluating the OMI-derived isoprene emissions with relaxed-eddy  
331 accumulation flux (REA) measurements obtained from above-canopy towers and aircraft during  
332 African field campaigns. OMI-derived isoprene emissions are on average 2 times higher than  
333 REA measurements over the equatorial forest and woody savannas but this could reflect biases in  
334 the measurements. MEGAN emissions are 5-10 times higher.

335 We subdivided Africa into 3 seasonally and ecologically coherent regions to examine the  
336 seasonality in OMI-derived isoprene emissions, and compare to the seasonality in MEGAN and  
337 in driving environmental variables. Equatorial forests exhibit weak seasonality that is driven  
338 predominantly by temperature, while seasonality in savannas is driven by both temperature and  
339 LAI, in a manner consistent with MEGAN.

340 Isoprene emissions in MEGAN are computed as the product of (1) an emission factor  $E_o$   
341 characteristic of the PFT, (2) the LAI, and (3) activity factors dependent on local environmental  
342 variables. We applied the LAI and MEGAN activity factors to our OMI-derived isoprene  
343 emissions to obtain emission factors representative of different PFTs. These agree well with the  
344 ensemble of leaf-level flux measurements in Africa and imply large downward corrections to  
345 MEGAN emission factors for equatorial forests and semi-arid vegetation. Such corrections are  
346 consistent with the latitudinal gradient of isoprene across West Africa measured in the AMMA  
347 field campaign.

348 The OMI-derived emission factors can be incorporated into the MEGAN formalism (Eq.  
349 (1)) to improve modeling of isoprene emissions in Africa. The resulting isoprene emissions for  
350 the continent are  $77 \text{ Tg C a}^{-1}$ , as compared to  $104 \text{ Tg C a}^{-1}$  in the standard MEGAN inventory.  
351 Most of the difference is over equatorial Africa. We conducted GEOS-Chem simulations with  
352 and without African isoprene emissions (using OMI-derived emission factors) to examine the  
353 impact on regional particulate matter (PM) and oxidants. The largest effect of isoprene emissions  
354 on surface  $\text{O}_3$  is over West Africa where  $\text{NO}_x$  is high, and the largest effect on PM is also over  
355 West Africa because of pre-existing high concentrations of primary PM from combustion.

356

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362

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560 **Tables**561 **Table 1a. Relaxed Eddy Accumulation (REA) isoprene flux measurements in Africa<sup>a</sup>**

Site <sup>b</sup>	Observation Period		Platform	$E_{\text{ISOP}}$	Land Cover <sup>d</sup>	Reference
	Date	Time		[10 <sup>12</sup> atoms C cm <sup>-2</sup> s <sup>-1</sup> ] <sup>c</sup>		
1	Mar 1996	12-13 LT	tower	3.5	Equatorial trees	Serça et al. (2001)
	Nov 1996	12-13 LT		1.4		
2	Nov-Dec 1996	9:30-11:30 LT	aircraft	1.5	Equatorial trees	Greenberg et al. (1999)
3	Nov-Dec 1996	9:30-11:30 LT	aircraft	0.91	Woody savanna	Greenberg et al. (1999)
4	Feb 2001	12-13 LT	tower	1.2	Savanna	Harley et al. (2003)

562 <sup>a</sup>Data used to evaluate OMI-derived and MEGAN isoprene emissions (Fig. 2).563 <sup>b</sup>See Fig. 1 for the location of each site.564 <sup>c</sup>Mean values at 12-15 local time.565 <sup>d</sup>MODIS IGBP land cover classification (Fig. 1).

566 Table 1b. Leaf-level isoprene flux measurements in Africa<sup>a</sup>

Site <sup>b</sup>	Observation Period	$E_o$ [mg C m <sup>-2</sup> h <sup>-1</sup> ]	Land Cover <sup>c</sup>	Reference
1	1996	3.6	Equatorial trees	Serça et al. (2001)
2	1995-1996	1.0	Equatorial trees	Klinger et al. (1998)
3	1995-1996	1.2	Woody savanna	Greenberg et al. (1999)
4	Feb 2001	0.47	Savanna	Harley et al. (2003)
	Dec 1992	2.4	Savanna	Guenther et al. (1996)
5	1995-1996	1.4	Woody savanna	Klinger et al. (1998)
6	1995-1996	3.0	Woody savanna	Klinger et al. (1998)
7	1995-1996	0.95	Savanna	Klinger et al. (1998)
8	Dec 1992	4.4	Savanna	Guenther et al. (1996)
	Feb-Mar 2001	4.5	Savanna	Otter et al. (2002)
9	Feb-Mar 2001	0.70	Shrubs	Otter et al. (2002)
10	Feb-Mar 2001	0.70	Savanna	Otter et al. (2002)
11	Feb-Mar 2001	8.2	Woody savanna	Otter et al. (2002)
	Feb-Mar 2001	3.6	Woody savanna	Otter et al. (2002)

567 <sup>a</sup>Emission factors  $E_o$  for standard conditions of temperature and PAR (Eq. (1)) used to compare to  
568 MEGAN and OMI-derived values in Fig. 5.

569 <sup>b</sup>See Fig. 1 for the location of each site.

570 <sup>c</sup>MODIS IGBP land cover classification (Fig. 1).

571

572 Table 2. Isoprene emission factors for African plant functional types<sup>a</sup>

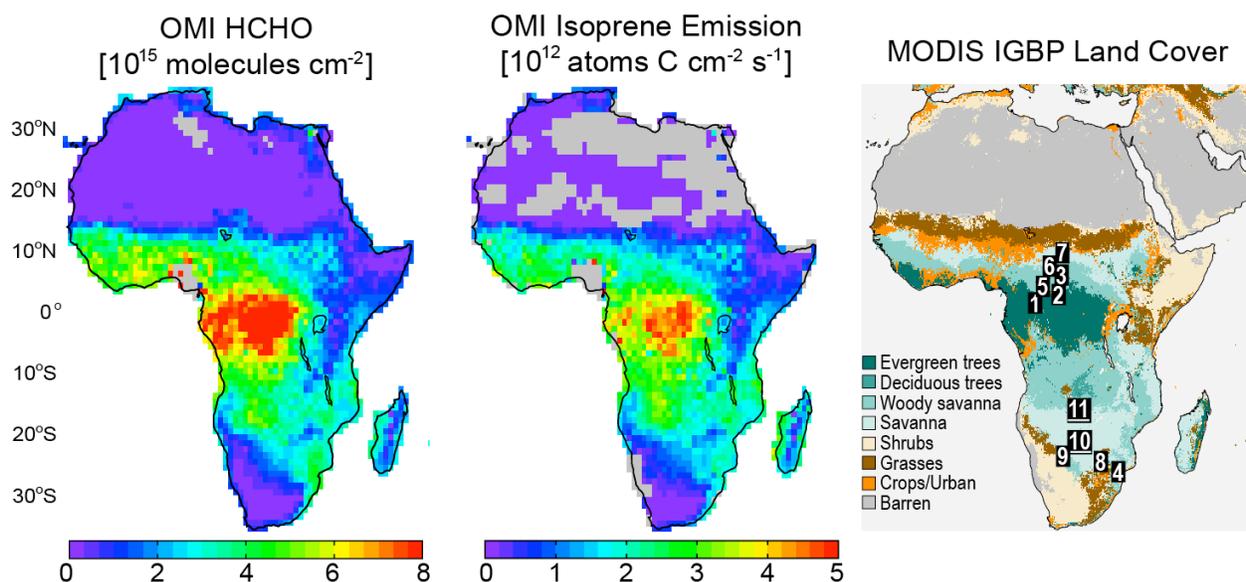
Plant functional type	Emission factor [mg C m <sup>-2</sup> h <sup>-1</sup> ]	
	MEGAN	OMI
MODIS IGBP classification <sup>b</sup>		
Evergreen broadleaf trees	4.3 ± 2.0	2.7 ± 1.0
Deciduous broadleaf trees	4.4 ± 1.7	2.9 ± 0.2
Woody savannas	3.2 ± 1.3	2.6 ± 1.0
Savannas	2.9 ± 1.2	2.3 ± 0.8
Shrubs	3.0 ± 1.3	1.6 ± 0.8
Grasses	1.8 ± 0.9	1.6 ± 0.9
Crops	1.4 ± 0.9	1.6 ± 0.6
Mosaic of crops and natural vegetation	2.3 ± 1.1	2.5 ± 1.0
GLC 2000 classification <sup>c</sup>		
Evergreen broadleaf trees	4.4 ± 1.9	2.5 ± 1.0
Deciduous broadleaf trees	3.0 ± 1.3	2.7 ± 0.9
Shrubs	3.1 ± 1.6	2.2 ± 1.0
Herbs	2.4 ± 1.3	1.9 ± 0.9
Sparse herbs or shrubs	2.4 ± 1.2	1.5 ± 0.7
Cultivated land	1.8 ± 1.1	2.2 ± 0.9
Mosaic of crops and natural vegetation	3.0 ± 1.3	2.7 ± 0.7
Mosaic of crops and shrubs or grasses	2.6 ± 1.0	1.9 ± 0.9

573 <sup>a</sup> Isoprene emission factor  $E_o$  in Eq. (1) at standard conditions of air temperature (303 K) and  
574 photosynthetically active radiation (1000  $\mu\text{mol photons m}^{-2} \text{s}^{-1}$ ). Values are means and standard  
575 deviations obtained by mapping the  $E_o$  data from Fig. 5 onto the MODIS IGBP and GLC 2000  
576 land maps. Plant functional type classifications are as given by each land map.

577 <sup>b</sup> Friedl et al. (2002) and shown in Fig. 1

578 <sup>c</sup> Mayaux et al. (2006)

579 **Figures**

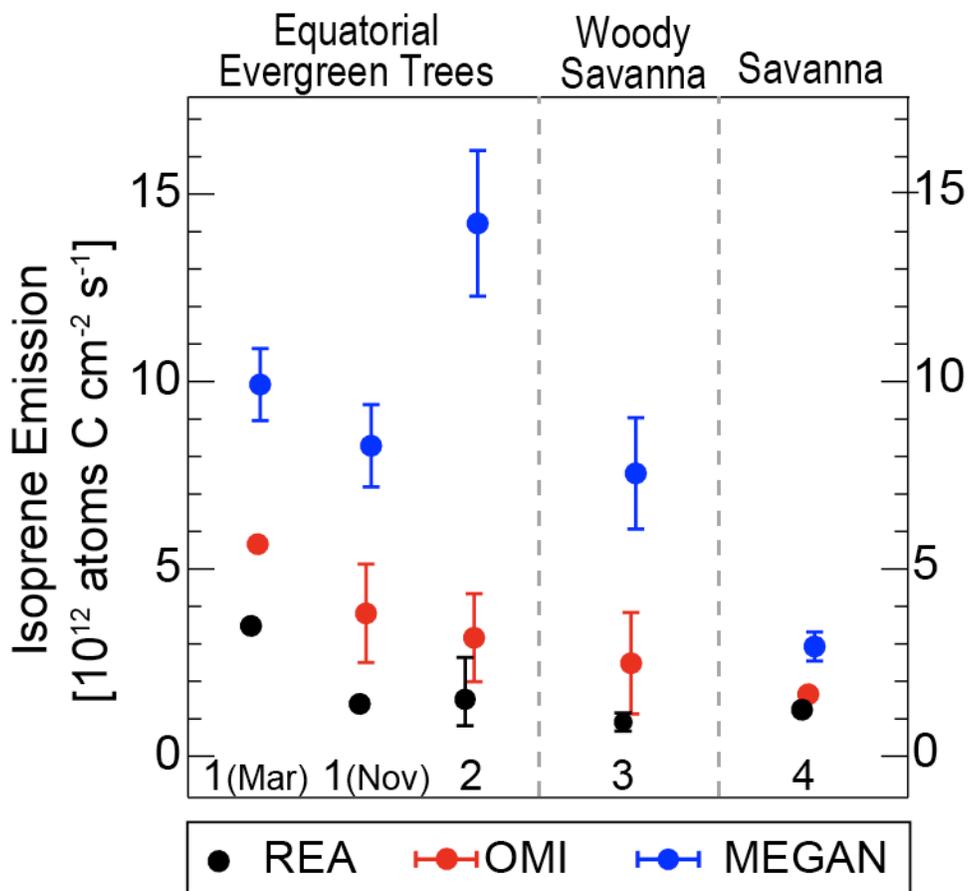


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582 **Figure 1.** Annual mean (2005-2009) OMI HCHO vertical columns at 1×1° horizontal resolution  
583 screened against biomass burning and anthropogenic HCHO (left), and resulting OMI-derived  
584 isoprene emissions (center), as derived by Marais et al. (2012) and summarized in the text. The  
585 OMI observations are at 13:30 local time (LT) and the OMI-derived isoprene emissions are for  
586 12-15 LT. The right panel is a MODIS IGBP land cover map (Friedl et al., 2002) with numbers  
587 showing the location of isoprene flux measurements used to evaluate the OMI-derived isoprene  
588 emissions (Tables 1a and 1b).

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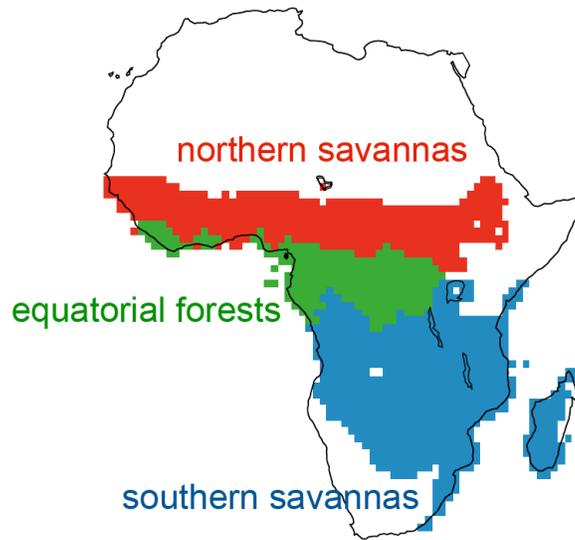


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592 **Figure 2.** Mean canopy-scale isoprene emissions at African sites 1-4 (see Fig. 1 and Table 1a)  
 593 measured by relaxed-eddy accumulation (REA), and comparison to OMI-derived and MEGAN  
 594 values. All values are for 12-15 local time, with diurnal correction for REA measurements at  
 595 sites 2 and 3 (see text). Vertical bars on the REA measurements for sites 2-3 are the interquartile  
 596 ranges over the aircraft sampling domain given in Greenberg et al. (1999). OMI-derived and  
 597 MEGAN values are 2005-2009 monthly averages for the site locations and observation times,  
 598 with interannual standard deviations shown as vertical bars. Mar=March, Nov=November.

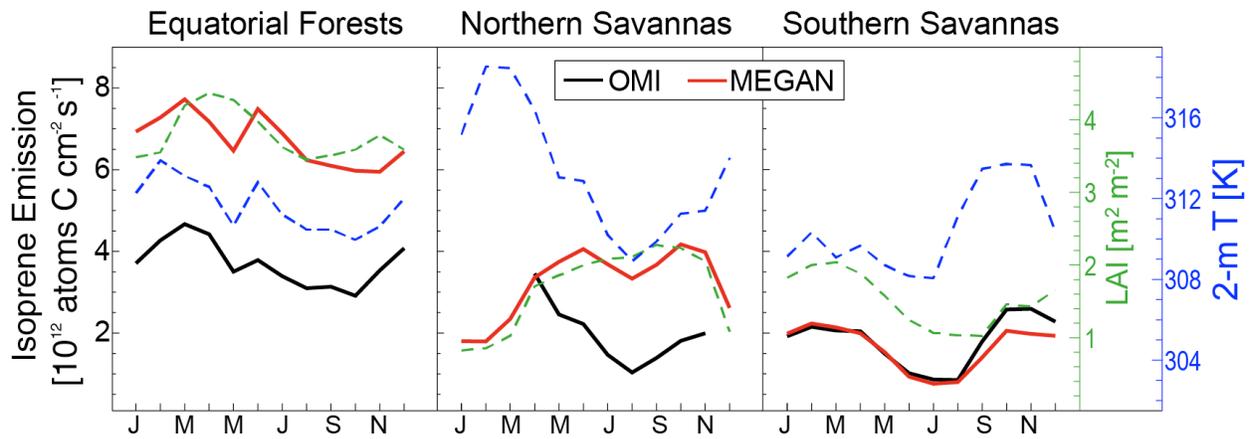
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601 **Figure 3.** Coherent regions used for analysis of the factors controlling OMI-derived isoprene  
602 emissions. Land cover definitions are from the MODIS IGBP map (Fig. 1).

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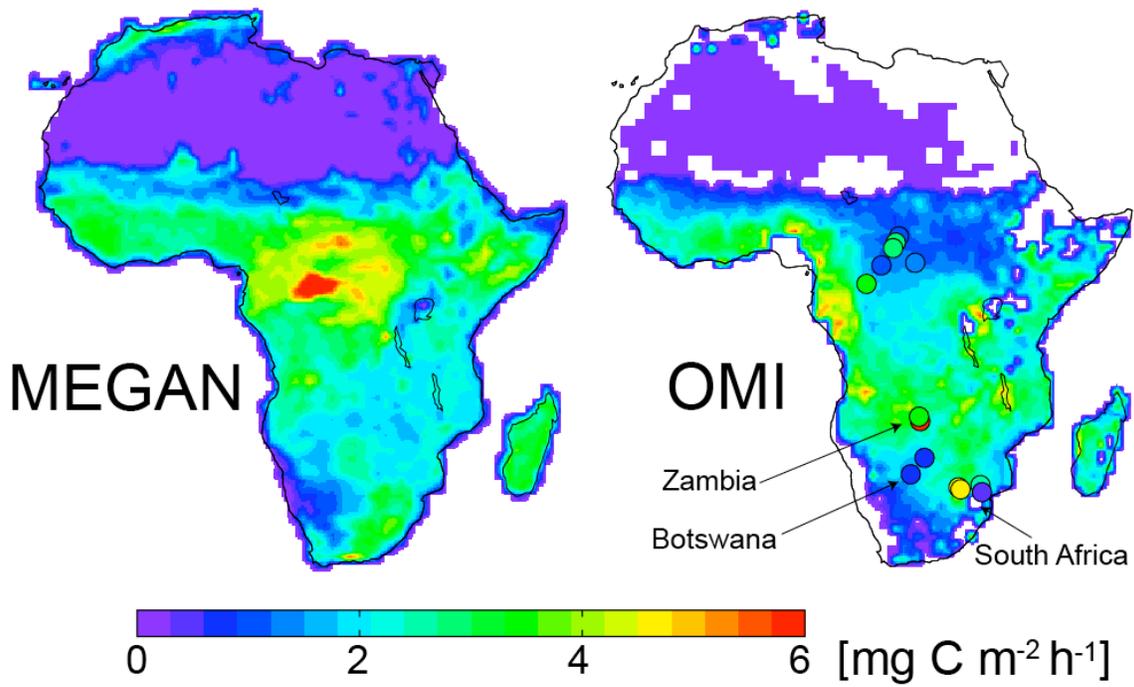


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606 **Figure 4.** Seasonality of isoprene emissions and environmental variables averaged over the  
 607 coherent African regions of Fig. 3. Monthly mean OMI-derived (black) and MEGAN (red)  
 608 isoprene emissions are shown together with GEOS-5 2-m temperature (blue) and MODIS LAI  
 609 (green). OMI-derived isoprene emissions are not available for northern savannas in December-  
 610 March because of biomass burning interference. Emissions and 2-m temperature are for 12-15  
 611 local time. LAI is the combined Terra and Aqua product (Yang et al., 2006). All data are means  
 612 for 2005-2009.

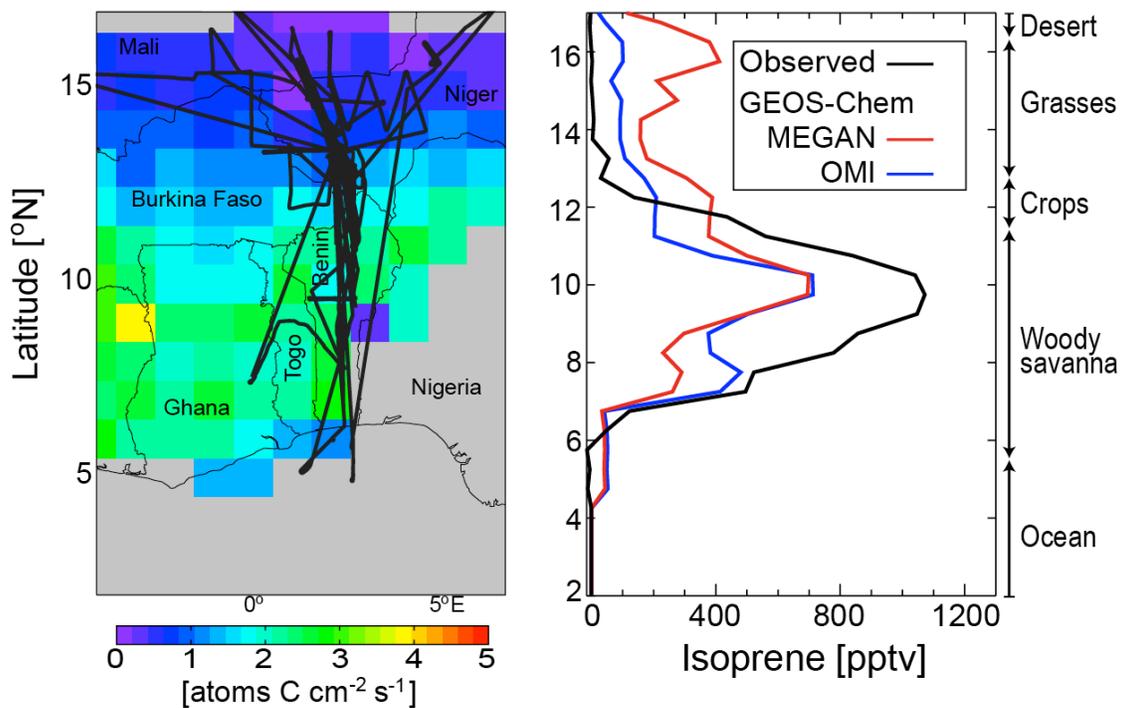
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615 **Figure 5.** Isoprene emission factors ( $E_o$  in Eq. (1)) representing the emission flux under standard  
 616 conditions. Measured landscape-level emission factors from field sites (circles; see Fig. 1 and  
 617 Table 1b) are compared to those used in MEGAN (left) and obtained with OMI (right). White  
 618 indicates missing OMI data.

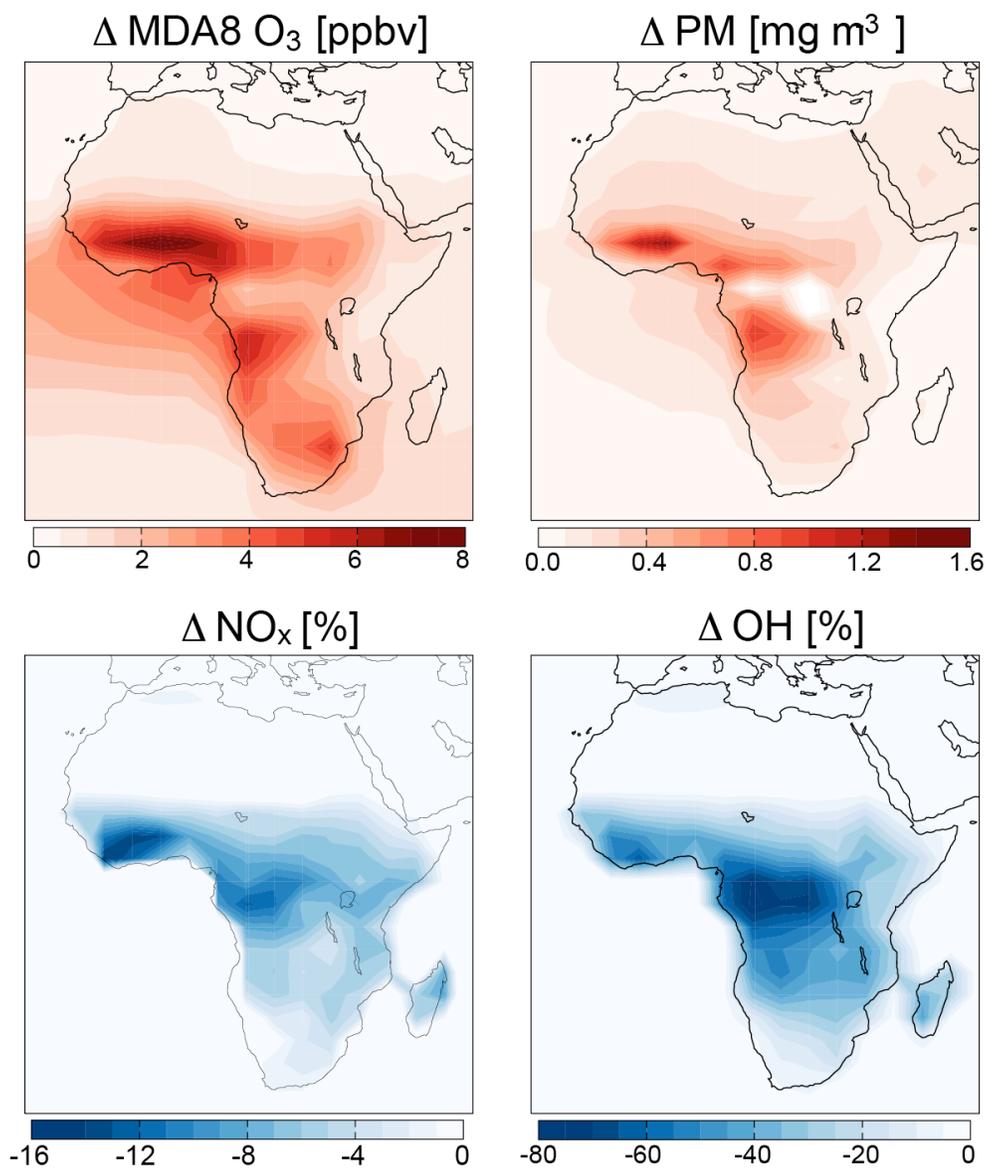
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621 **Figure 6.** Latitudinal variability of isoprene in West Africa. Left panel shows the July-August  
 622 2006 AMMA flight tracks superimposed on July-August 2005-2009 OMI-derived isoprene  
 623 emissions (Marais et al., 2012). Right panel shows boundary-layer (< 1 km) isoprene  
 624 concentrations along the AMMA flight tracks; observations averaged over 0.5° latitude bands are  
 625 compared to GEOS-Chem model results using either MEGAN or OMI-derived isoprene  
 626 emission factors  $E_o$ . Dominant MODIS IGBP biomes along the flight tracks are indicated.

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630 **Figure 7.** Effect of African isoprene emissions on regional oxidant and particulate matter (PM)  
 631 concentrations in surface air. Shown are the annual mean differences  $\Delta$  between GEOS-Chem  
 632 simulations with and without African isoprene emissions. Isoprene emission is computed using  
 633 OMI-derived emission factors.

634