

1 **Spatial and temporal variability in the ratio of trace gases emitted from biomass**  
2 **burning**

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9

10 **Abstract**

11 Fires are a major source of trace gases and aerosols to the atmosphere. The amount of  
12 biomass burned is becoming better known, most importantly due to improved burned area  
13 datasets and a better representation of fuel consumption. The spatial and temporal variability  
14 in the partitioning of biomass burned into emitted trace gases and aerosols, however, has  
15 received relatively little attention. To convert estimates of biomass burned to trace gas and  
16 aerosol emissions, most studies have used emission ratios (or emission factors [EFs]) based  
17 on the arithmetic mean of field measurement outcomes, stratified by biome. However, EFs  
18 vary substantially in time and space, even within a single biome. In addition, it is unknown  
19 whether the available field measurement locations provide a representative sample for the  
20 various biomes. Here we used the available body of EF literature in combination with  
21 satellite-derived information on vegetation characteristics and climatic conditions to better  
22 understand the spatio-temporal variability in EFs. While focusing on CO, CH<sub>4</sub>, and CO<sub>2</sub>, our  
23 findings are also applicable to other trace gases and aerosols. We explored relations between  
24 EFs and different measurements of environmental variables that may correlate with part of the  
25 variability in EFs (tree cover density, vegetation greenness, temperature, precipitation, and the  
26 length of the dry season). Although reasonable correlations were found for specific case  
27 studies, correlations based on the full suite of available measurements were lower ( $r$ -  
28 max=0.62). This may be partly due to uncertainties in the remotely sensed data, differences in  
29 measurement techniques for EF, assumptions on the ratio between flaming and smoldering  
30 combustion, and incomplete information on the location and timing of EF measurement. We

31 derived new mean EFs, using the relative importance of each measurement location. These  
32 weighted averages were relatively similar to the arithmetic mean. When using relations  
33 between the environmental variables and EFs to extrapolate to regional scales, we found  
34 substantial differences, with for example a ~19% lower CO EF for savannas and grasslands in  
35 Australia compared to southern hemisphere South America. We argue that from a global  
36 modeling perspective, future measurement campaigns could be more beneficial if  
37 measurements are made over the full fire season, and if relations between ambient conditions  
38 and EFs receive more attention.

39

## 40 **1 Introduction**

41 Although biomass burning is one of the most ancient forms of anthropogenic atmospheric  
42 pollution, its importance for atmospheric chemistry has only been recognized since the late  
43 seventies (Radke et al., 1978; Crutzen et al., 1979). Interest in this topic grew when studies  
44 suggested that for several trace gases and aerosol species, biomass burning emissions could  
45 rival fossil fuel emissions (Seiler and Crutzen, 1980; Crutzen and Andreae, 1990), and that  
46 these vegetation fires could affect large parts of the world due to long-range transport  
47 processes (Andreae, 1983; Fishman et al., 1990; Gloudemans et al., 2006). During the last two  
48 decades biomass burning has received considerable interest, leading for example to the  
49 realization that vegetation fires impact 8 out of 14 identified radiative forcing terms (Bowman  
50 et al., 2009), contribute to interannual variability (IAV) in growth rates of many trace gases  
51 (Langenfelds et al., 2002), and influence human health and plant productivity downwind of  
52 fires through enhanced ozone and aerosol concentrations (e.g. Sitch et al., 2007).

53 To assess the atmospheric impact of biomass burning quantitatively, accurate data on the  
54 emission of trace gases and aerosols is required. Crucial parameters include burned area, fuel  
55 consumption, and the emission factor (EF), usually defined as the amount of gas or particle  
56 mass emitted per kg of dry fuel burned, expressed in units of g/kg dry matter (DM) (Andreae  
57 and Merlet, 2001).

58 Pioneering experiments to characterize fire emissions were conducted in South America  
59 (Crutzen et al., 1979), Africa (Delmas, 1982), and Australia (Ayers and Gillett, 1988). In the  
60 beginning of the 1990s, the experiments of these individual groups were followed by a  
61 number of large international biomass burning experiments in various ecosystems throughout  
62 the world. These included the Southern Africa Fire-Atmosphere Research Initiative (SAFARI

63 92 and SAFARI 2000) in southern Africa (Lindesay et al., 1996; Swap et al., 2002),  
64 Dynamique et Chimie Atmosphérique en Forêt Equatoriale-Fire of Savannas (DECAFE-FOS)  
65 in West Africa (Lacaux et al., 1995), Transport and Atmospheric Chemistry Near the Equator-  
66 Atlantic (Trace-A) over Brazil, southern Africa, and the South Atlantic (Fishman et al., 1996),  
67 Fire Research Campaign Asia-North (FireSCAN) in central Siberia (FIRESCAN Science  
68 Team, 1996), and Smoke, Clouds, and Radiation-Brazil (SCAR-B) in Brazil (Kaufman et al.,  
69 1998).

70 These coordinated studies and numerous independent smaller investigations have resulted in a  
71 large body of information on emission characteristics. Several summaries of experimental EF  
72 data were given (e.g. Andreae, 1993; Delmas et al., 1995; Akagi et al., 2010). The most  
73 extensive and frequently used summary is given by Andreae and Merlet (2001), in which all  
74 the available data on fire emission characteristics for a large number of chemical species was  
75 synthesized into a consistent set of units. The measurements were stratified by biome type or  
76 fire use; tropical forest fires (in general fires used in the deforestation process), savanna and  
77 grassland fires, extratropical forest fires, biofuel burning, charcoal making, charcoal burning,  
78 and the burning of agricultural residues. The database is updated annually (Andreae, personal  
79 communication, 2009) and we will refer to this as A&M2001-2009 in the remainder of this  
80 paper.

81 Including fire processes in dynamic global vegetation models (DGVM) and biogeochemical  
82 models led to a better understanding of the spatio-temporal variability in fuel loads and fire  
83 processes. For example, annual global burned area estimates (Giglio et al., 2006; Giglio et al.,  
84 2010) and global emissions estimates according to the Global Fire Emissions Database  
85 (GFED; van der Werf et al., 2006; van der Werf et al., 2010) are decoupled on an annual  
86 timescale because most burned area occurs in savanna-type ecosystems with relatively low  
87 fuel loads, while the smaller areas that burn in forested ecosystems results in higher emissions  
88 per unit area burned due to fuel loads that are at least one order of magnitude larger.

89 New burned area products (L3JRC [Tansey et al., 2007], MODIS [Roy et al., 2008; Giglio et  
90 al., 2010], GLOBCARBON [Plummer et al., 2006]) allow for a better characterization of the  
91 timing and locations of fire, although the quality of these burned area products varies and they  
92 may have difficulties in capturing small fires (Chang et al., 2009; Roy and Boschetti, 2009;  
93 Giglio et al., 2010). When accounting for errors in transport and chemistry as well as  
94 uncertainties in satellite retrievals of trace gases and aerosols, combining bottom-up (such as

95 GFED) and top-down methods potentially allows for an assessment of the magnitude of  
96 emissions as well as their spatio-temporal variability (Arellano et al., 2004; Edwards et al.,  
97 2004; Gloude-mans et al., 2006). This requires a thorough understanding of the relations  
98 between biomass combusted and emission of the trace gases or aerosols that are used as top-  
99 down constrains, most often CO.

100 Although our knowledge on the spatial and temporal variability of fire substantially increased  
101 in the last decade due to new satellite information, the total amount of biomass combusted,  
102 and especially the partitioning of combusted carbon (C) into different combustion products, is  
103 improving but still uncertain. To date, most large-scale studies have used the average EFs  
104 provided by A&M2001-2009. EFs, however, show large variability, mainly due to differences  
105 in fuel type and composition, burning conditions, and location (Andreae and Merlet, 2001;  
106 Korontzi et al., 2003). Even though EFs may vary in time and space, this variability is usually  
107 not taken into account in large-scale emissions assessments except for variations due to  
108 vegetation type (in general all savanna fires, all tropical forest fires, all extratropical forest  
109 fires, and all agricultural waste burning fires have their own, averaged, EFs). In addition to  
110 the lack of representation in spatio-temporal variability, the often-used average EFs may have  
111 limitations because it is not known whether they are based on a representative sample of a  
112 specific vegetation type.

113 In the literature only a few papers on regional emissions estimates considered seasonal and/or  
114 spatial variability of EFs into account. Hoffa et al. (1999) related fire emissions in Zambian  
115 grasslands and woodlands with PGREEN, defined as the proportion of green grass biomass to  
116 total (green+dead) grass biomass. Ito & Penner (2005) applied three different EF scenarios  
117 that accounted for both seasonal and spatial variability. Both studies confirmed that a spatial  
118 and temporal varying EF can have a significant impact on regional emissions estimates.

119 Here we evaluated existing information on EFs, based on an extensive database of field  
120 measurements (A&M2001-2009), and systematically explored several environmental  
121 variables that may be related to the spatial and temporal variability in EFs. Data on fraction  
122 tree cover, precipitation, temperature, Normalized Difference Vegetation index (NDVI, a  
123 measure of vegetation greenness or productivity), and length of the dry season were used to  
124 develop relations with the EFs for different vegetations types. We focused on CO, methane  
125 (CH<sub>4</sub>), and CO<sub>2</sub>. However, since the Modified Combustion Efficiency (MCE, defined as the  
126 amount of C released as CO<sub>2</sub> divided by the amount of C released as CO<sub>2</sub> plus CO (Yokelson

127 et al., 1996)) has been used as an effective predictor for the emission of smoke gas  
128 composition from biomass fires (e.g., Ward et al., 1996; Sinha et al., 2003; Yokelson et al.,  
129 2003) and for certain aerosol species and characteristics (e.g., Christian et al., 2003;  
130 McMeeking et al., 2009; Janhäll et al., 2010), our findings on CO and CO<sub>2</sub> EFs can be used to  
131 better understand emissions of other trace gases and aerosols as well. We restricted our  
132 analysis to in-situ measurements due to the focus on spatio-temporal variability as a result of  
133 variability in vegetation and climatic conditions; laboratory measurements of EFs were not  
134 taken into account. We present new weighted EFs for specific vegetation types, and indicate  
135 how future EF experiments could be more beneficial from a global modeling perspective.

136

## 137 **2 Fire processes**

138 To facilitate the description of the main factors that influence the EF of different trace gases  
139 (section 2.2), we start with a brief summary of the combustion process (section 2.1). For more  
140 detailed information the reader is referred to Chandler et al. (1983), Lobert and Warnatz  
141 (1993), and Yokelson et al. (1996; 1997).

### 142 **2.1 The combustion process**

143 The combustion of the individual fuel elements proceeds through a sequence of stages  
144 (ignition, flaming, smoldering, and extinction), each with different chemical and physical  
145 processes that result in different emissions.

146 The initial ignition is the phase before a self-sustaining fire can start, and it depends on both  
147 fuel (size, density, water content) and environmental (temperature, relative humidity, wind)  
148 factors whether the fuel is ignited or not. Once the fuel is sufficiently dry, combustion can  
149 proceed from the ignition phase to the flaming phase. It starts with thermal degradation, in  
150 which water and volatile contents of the fuel are released, and is followed by the thermal  
151 cracking of the fuel molecules (pyrolytic step); high-molecular compounds are decomposed to  
152 char (less volatile solids with high C content), tar (molecules of intermediate molecular  
153 weight), and volatile compounds. When diluted with air, a flammable mixture may form.  
154 Many different compounds are produced during this phase, particularly CO<sub>2</sub> and H<sub>2</sub>O.

155 After most volatiles have been released and the rate of the pyrolysis slows down, less  
156 flammable compounds are produced; the flaming combustion ceases, and the smoldering

157 phase begins. Smoldering combustion is a lower-temperature process compared to flaming  
158 combustion emitting large amounts of incompletely oxidized compounds (e.g. CO), and can  
159 proceed for days, even under relatively high moisture conditions. The slower rate of pyrolysis  
160 results in lower heat production and therefore in a lower decomposition rate, until the process  
161 terminates (extinction phase). The most common causes of extinction are a physical gap in the  
162 fuels that prevents sufficient heat transfer to additional fuels, rainfall, or fire spread into wet  
163 fuels.

164 The combustion processes described above are somewhat simplified, and in most fires all of  
165 these processes occur simultaneously in different parts of the fuel bed. For real-time open  
166 vegetation fires, different factors that influence the combustion process and which may  
167 change over time (e.g. meteorological conditions, differences in aboveground biomass  
168 density, topography) also need to be considered. The amount of substances emitted from a  
169 given fire and their relative proportions are determined to a large extent by the ratio of  
170 flaming to smoldering combustion, which is related to the combustion efficiency (CE),  
171 defined as the fraction of the fuel C burned converted to CO<sub>2</sub>.

172

## 173 **2.2 Factors influencing the EF**

174 The exact physical relations between environmental variables and EFs are not well  
175 understood, although recent laboratory studies have aimed to quantify how, for example,  
176 moisture content impacts EFs (e.g. Chen et al., 2010). Qualitatively, important parameters that  
177 partly govern the flaming / smoldering ratio and thus EFs include vegetation characteristics,  
178 climate, weather, topography, and fire practices.

179 A variable that may affect both the behavior and the emissions of a fire is the water content of  
180 the vegetation. The water content partly determines whether a plant or tree can ignite and  
181 what the combustion efficiency will be. Water in plants or trees has the capability to either  
182 stop a fire completely or to slow down the burning process (to a low smoldering stage).  
183 However, also wet fuels can ignite if a sustained ignition source is applied. For instance,  
184 crown fires spread at high rates with large flames burning fresh foliage with high moisture  
185 content.

186 Other fuel characteristics related to vegetation are the size, density, and the spacing of the  
187 fuels. Some studies (Bertschi et al., 2003; McMeeking et al., 2009) suggest that combustion

188 completeness, defined as the fraction of biomass exposed to a fire that was actually consumed  
189 (or volatilized) in a fire, is impacted more by fuel spacing than fuel moisture. It is likely fuel  
190 spacing is equally important in driving variability in EFs. Because fuel has to be heated to  
191 ignition temperature, small low-density fuel particles are more easily ignited than larger high-  
192 density particles. Once burning, the rate of heat production for smaller particles is higher than  
193 for larger particles, and therefore smaller particles are also capable of sustaining flaming  
194 combustion and supporting the burning of larger particles. In general, grass fuels in savannas  
195 have a large surface to volume ratio, are more easily pyrolyzed, and therefore burn largely in  
196 the flaming phase, while stems and coarse litter that burn in forest fires are not as well  
197 oxidized and burn more in the smoldering phase. However, with an efficient heat transfer  
198 between fuel elements even large logs in deforestation fires can be consumed mostly by  
199 flaming combustion (Christian et al., 2007; McMeeking et al., 2009).

200 Climate also plays an important role in the existence and settlement of vegetation, and thus  
201 determines the availability of fire fuel (Lobert and Warnatz, 1993). Fire frequency and the fire  
202 season are also partly determined by climatic factors. Weather has a more short-term impact  
203 on fire. Temperature, precipitation, and wind speed are factors that partly determine the  
204 occurrence of fires as well as their behavior, especially the CE. Temperature may affect the  
205 fire probability and ignition due to its effect on fuel moisture. Precipitation is capable of  
206 inhibiting, completely stopping, or preventing a fire. Wind can have an effect on the spread  
207 rate of a fire, as fires usually propagate in two different directions; with the wind (heading  
208 fires) and into the wind (backing fires). The local topography can also change the burning  
209 behavior of a fire; heat rises and an upslope fire therefore achieves better heat transfer from  
210 the burning fuels to the unburned fuels. If all other conditions are equal, this leads to fires that  
211 spread faster.

212 In the tropics and subtropics, fire is mainly a human-driven process. We expect that regional  
213 variations in fire practices influences EFs, especially in agricultural fires and fires used in the  
214 deforestation process. Slash and burn fires, for example, are different from the burning of  
215 fuels that have been mechanically piled together into windrows and may burn more intense.  
216 This practice requires heavy machinery and is therefore limited to regions with more capital,  
217 for example the southern part of the Amazon where forests are cleared for soy production,  
218 amongst others (Morton et al., 2006).

219 In summary, both the combustion process and its inter-relationship with the environment are  
220 very complicated. At present, literature focusing on how environmental variables impact EFs  
221 from real fires is limited and data from laboratory studies is often conflicting and  
222 inconclusive. Nevertheless, empirical relationships between satellite observables and EF may  
223 exist and are further explored here.

224

### 225 **3 Literature database of EF measurements**

#### 226 **3.1 Introduction**

227 We used the EF database for different vegetation types that was compiled by A&M2001-  
228 2009. The database consists of EFs measured during individual experiments, as well as during  
229 large international measurement campaigns. The database includes both field data (sampled  
230 on the ground or from aircraft) and laboratory measurements. We excluded laboratory  
231 measurements in our analyses because the focus of our work is on EF variability and the role  
232 of local (climatic) conditions, which are better represented by EF measurements in the field.  
233 In addition, laboratory measurements may not be fully representative of burning conditions in  
234 the field; it is for example impractical to burn a diverse suite of large diameter tropical logs in  
235 the lab (Yokelson et al., 2008). In the work of A&M2001-2009, laboratory measurements  
236 were also excluded for calculating biome-averaged EFs for CO, CH<sub>4</sub>, and CO<sub>2</sub>.

237 Most of the EFs in the database of A&M2001-2009 are measured using the C mass balance  
238 (CMB) method (Ward et al., 1979; Radke et al., 1990). The underlying premise of this  
239 method is that all C combusted in a fire is emitted into measurable portions in five forms:  
240 CO<sub>2</sub>, CO, CH<sub>4</sub>, non-methane hydrocarbons (NMHC), and particulate C in smoke particles.  
241 The EF of a species is then calculated from the ratio of the mass concentration of those  
242 species to the total carbon concentration emitted in the plume. To convert the EF to g/kg DM  
243 of fuel burned, the data need to be multiplied with the carbon content of the fuel. A&M2001-  
244 2009 adopted a C content of 45% when this information was not given in literature cited.  
245 However, a detailed study of Susott et al. (1996) suggests a global average C fraction for  
246 biomass closer to 50%, with a considerable range, which would indicate an additional ~10%  
247 uncertainty in addition to other uncertainties.

248 When the emission data were given as molar emission ratios, A&M2001-2009 used the  
249 molecular weights of the trace and reference species to calculate the EF. Molar emission

250 ratios can be obtained by dividing excess trace species concentrations measured in a fire  
251 plume by the excess concentration of a simultaneously measured reference gas (most often  
252 CO<sub>2</sub>). If the EF of the reference species was not provided, the mean EF for the specific type of  
253 fire was used.

254 With the A&M2001-2009 database as a starting point, we compiled all EFs and searched the  
255 literature for accompanying ancillary data such as measurement location and timing. We then  
256 expanded the database to include location-specific parameters related to vegetation type and  
257 climate of each measurement. We focused on the EFs of CO<sub>2</sub>, CO, and CH<sub>4</sub> because these  
258 gases were measured during most campaigns, and the EF of CO<sub>2</sub> and CO can be used to  
259 calculate the modified combustion efficiency (MCE), which can be used to predict EFs of  
260 other species (e.g., Ward et al., 1996; Sinha et al., 2003; Yokelson et al., 2003).

261

### 262 **3.2 Available EF data**

263 Figure 1 provides an overview of the locations where ground- and aircraft EF measurements  
264 were conducted for CO and CO<sub>2</sub>, with a background of mean annual fire C emissions. Fire  
265 emissions were taken from the Global Fire Emission Database (GFED) version 3.1 (Giglio et  
266 al., 2010; van der Werf et al., 2010). GFED consists of 0.5°×0.5° gridded monthly  
267 parameters; burned area, fuel loads, combustion completeness, and fire C losses. Fire  
268 emissions were estimated based on burned area (Giglio et al., 2010) in combination with the  
269 Carnegie-Ames-Stanford Approach (CASA) biogeochemical model to calculate fuel  
270 consumption. See van der Werf et al. (2010) for more information.

271 Most locations with both CO and CO<sub>2</sub> EF measurements are in North America, the arc of  
272 deforestation in the Brazilian Amazon, southern Africa (South Africa and Zambia), and  
273 northern Australia (Figure 1). While these areas are all major biomass burning regions,  
274 several other important regions lack measurements. These include Central Africa (e.g. Congo,  
275 Angola, but also regions further north such as Chad and southern Sudan), Siberia, Indochina,  
276 and Indonesia, although laboratory studies for Indonesian fuel samples exist (Christian et al.,  
277 2003). Most of these missing regions likely has relatively high rates of emissions of reduced  
278 gases compared to sampled regions; more woodland burning in Central Africa compared to  
279 southern Africa where most savanna measurements were made, more groundfires in boreal  
280 Asia compared to boreal North America where most extratropical EFs were measured, and  
281 moister conditions and more peat burning in Indonesia compared to South America where

282 most deforestation fire EFs were made. On the other hand, most measurements in Australia  
283 were made in the relatively moist part in the North while fires burning in the more arid  
284 interior have not been sampled.

285 To highlight the large variability in EFs, we plotted CH<sub>4</sub> EFs against the molar MCE (based  
286 on CO and CO<sub>2</sub> EFs) in Figure 2 for three different biomes. The biome-averaged EF values of  
287 A&M2001-2009 are also shown. In general, EFs in savannas & grasslands show high MCEs  
288 and a relatively low EF for CH<sub>4</sub>, mainly because burning mostly takes place in the flaming  
289 phase. Tropical forest measurements on the other hand, show lower MCEs and higher values  
290 for the EF of CH<sub>4</sub>, because these fires burn predominantly in the smoldering phase. This is  
291 also the case for the extratropical forest measurements, although here the values are more  
292 variable. The correlation coefficient (*r*) between MCE and CH<sub>4</sub> for all these in-situ  
293 measurements was -0.71 ( $EF_{(CH_4)} = -85.889 \times MCE + 85.278$ ), and correlation coefficients for  
294 the different vegetation types were -0.80 ( $EF_{(CH_4)} = -61.447 \times MCE + 61.142$ ), -0.81 ( $EF_{(CH_4)}$   
295  $= -104.551 \times MCE + 104.590$ ), and -0.52 ( $EF_{(CH_4)} = -59.992 \times MCE + 60.967$ ) for savanna and  
296 grasslands, tropical forest, and extratropical forest, respectively. Two extratropical forest  
297 measurements (Cofer et al., 1998: MCE=0.78, EF CH<sub>4</sub> = 4.5; Hobbs et al., 1996: MCE=0.81,  
298 EF CH<sub>4</sub>=16.2) were excluded from this graph for clarity, but they were taken into account to  
299 calculate the correlation coefficient.

300 Although lowering the number of EF studies in general decreases the correlation coefficient,  
301 several individual studies focusing on a selected number of measurements found higher  
302 correlation coefficients than the ones reported above. Yokelson et al. (2003) found a  
303 correlation coefficient of -0.93 ( $EF_{(CH_4)} = -48.522 \times MCE + 47.801$ ) for 8 African savanna  
304 fires. Korontzi et al. (2003) also found higher correlations and a slightly different slope for the  
305 regression of southern African savanna measurements - grasslands had a correlation of 0.94  
306 ( $EF_{(CH_4)} = -43.63 \times MCE + 42.951$ ) and for woodlands a correlation of 0.98 ( $EF_{(CH_4)} = -$   
307  $58.214 \times MCE + 56.710$ ) was found. Both vegetation types combined gave an overall  
308 correlation of 0.94, and a trendline of  $EF_{(CH_4)} = -47.948 \times MCE + 47.068$ .

309 For the tropical forest biome, Yokelson et al. (2008) found a correlation coefficient of 0.72 for  
310 9 fire-averaged MCEs and CH<sub>4</sub> EFs. The slope of this regression was significantly more  
311 gentle ( $EF_{(CH_4)} = -47.105 \times MCE + 48.555$ ) than the slope for this biome using all  
312 measurements in the A&M2001-2009 database. In older work, comparisons between the CE  
313 (which correlates well with the MCE) and CH<sub>4</sub> EFs was presented. Ward et al. (1992) showed

314 a correlation of 0.96 and a slope of  $EF_{(CH_4)} = -82.1 \times CE + 78.6$  for a regression of 18  
315 deforestation fires in Brazil. We are not aware of any recent comparisons between MCE and  
316 EF CH<sub>4</sub> for fires in the extratropical forest biome, but in older work of e.g. Ward & Hardy  
317 (1991) and Hao and Ward (1993), an overall higher correlation ( $r > 0.8$ ) is found for  
318 extratropical forest measurements. The slope of the regression lines of these individual studies  
319 was more gentle than the slope we found for the whole dataset. Lab experiments (Christian et  
320 al., 2003; McMeeking et al., 2009; Burling et al., 2010) also show overall higher correlations  
321 between MCE and EF CH<sub>4</sub> than our results for all data for the different vegetation biomes  
322 combined.

323 Overall, higher correlation coefficients and flatter slopes for the EF CH<sub>4</sub> and MCE  
324 relationship were found for individual studies focusing on a relatively small number of EF  
325 measurements, compared to the whole EF database of A&M2001-2009. Possible  
326 explanations for these differences between the whole dataset compared to individual studies  
327 are discussed in section 4. Individual studies (e.g. Hao and Ward, 1993) have shown that the  
328 linear relationships between the MCE and EF of CH<sub>4</sub> are quite different for individual  
329 biomes, for reasons not fully understood. This is also apparent from Figure 2; the slope and  
330 intercept of the savanna and extropical forest biome compare very well, but the regression  
331 line of CH<sub>4</sub> EFs and their MCE derived for tropical forest biome shows a steeper slope and  
332 larger intercept. Most variation and therefore lower overall correlation coefficient was caused  
333 by the extratropical forest measurements.

334 The large variability (even within biomes) apparent from Figure 2 may be partly explained by  
335 the different environmental variables that we described in section 2.2. One is related to the  
336 timing of the measurement, and thus to weather conditions during the fire (e.g., Korontzi et  
337 al., 2003). Fires in savannas and tropical forest areas usually burn during the late dry season,  
338 when fuel moisture is in general at minimum. Prescribed burning in tropical savannas on the  
339 other hand is often exercised in the early part of the dry season, and is commonly advocated  
340 when fire is used as a land management tool. Early season burns are less intense and result in  
341 a smaller amount of vegetation consumed per unit area and –probably more important- lead to  
342 less damage to the soil compared to late season fires. Pastoralists burn extensively in the early  
343 dry season to stimulate regrowth of palatable grasses for their cattle; fire is used for rapid  
344 nutrient release prior to the new growing season by farmers, and early burning is used in

345 national parks as a preventive measure against late dry season fires which tend to have higher  
346 intensities and are in general more destructive (Frost, 1996; Williams et al., 1998).

347 We explored the seasonal variation of the fire emissions for all EF data where a detailed  
348 description of the location and date of measurements was provided. To investigate whether  
349 the available measurements captured the fire seasonality we compared the number of EF  
350 measurements conducted in a specific biome with the seasonal variation in C emissions  
351 according to GFED3.1 (Figure 3). Only the  $0.5^{\circ} \times 0.5^{\circ}$  grid cells enclosing the locations where  
352 EF measurements were conducted for CO, CH<sub>4</sub>, and CO<sub>2</sub> were used, and the seasonal cycle in  
353 each grid cell was normalized to its peak fire month (PFM). Figure 3a shows the seasonality  
354 of the number of EF measurements and the GFED3.1 fire emissions for all the EF  
355 measurement locations in the savanna and grassland biome for the PFM, and the months  
356 before and after the PFM. Results for the tropical forest biome are shown in Figure 3b.

357 For EF measurement locations in the savanna biome, 46% of the total annual amount of C  
358 was emitted by fires in the PFM, and 78% when also including the month before and after the  
359 PFM. For the tropical forest biome, this was 66% and 84%, respectively. The percentage of  
360 EF measurements conducted in the PFM was 23% for both the savanna and tropical forest  
361 biome, and respectively 71% and 88% when also including the month before and after the  
362 PFM. In other words, the current body of measurements have undersampled the peak fire  
363 month with especially the tropical forest fire measurements sampling earlier than desirable.  
364 Extratropical forest measurements were excluded from this analysis, because the fire season is  
365 much more variable from year to year compared to the tropics (Giglio et al., 2006).

366

### 367 **3.3 Remotely sensed environmental data**

368 One of our main objectives was to model the variability in CO, CH<sub>4</sub>, and CO<sub>2</sub> EFs. For this,  
369 we compared all the EFs in the database with global monthly datasets of potentially relevant  
370 parameters (as described in section 2.2); fraction tree cover, precipitation, temperature, NDVI,  
371 and the length of the dry season. These parameters were chosen since globally consistent  
372 information is available for a longer period of time, although the spatial and temporal  
373 resolution is relative coarse (typically  $0.5^{\circ} \times 0.5^{\circ}$  and monthly data) and may not fully capture  
374 key regional variability. Specific local and regional factors that may have a large influence on  
375 the EF variability, like e.g. wind, were excluded due to a lack of reliable data.

376 We used the fraction tree cover (FTC) product regridded to  $0.5^{\circ} \times 0.5^{\circ}$  resolution for the year  
377 2002 to represent the vegetation density and the ratio between herbaceous and woody fuels in  
378 the EF measurement locations. In the GFED modeling framework, FTC is the key control on  
379 the fraction coarse fuels that burn predominantly in the smoldering phase (e.g., stems, coarse  
380 woody debris) as opposed to fine fuels burning mostly in the flaming phase (leaves, grass,  
381 fine litter) in a grid cell. The FTC product was derived from the Vegetation Continuous Fields  
382 (VCF) collection which contains proportional estimates for vegetative cover types: woody  
383 vegetation, herbaceous vegetation, and bare ground (Hansen et al., 2003). The product was  
384 derived from seven bands of the MODerate-resolution Imaging Spectroradiometer (MODIS)  
385 sensor onboard NASA's Terra satellite. The continuous classification scheme of the VCF  
386 product better captures areas of heterogeneous land cover than traditional discrete  
387 classification schemes.

388 The  $1^{\circ} \times 1^{\circ}$  daily (1DD) Global Precipitation Climatology Project (GPCP) precipitation  
389 product (Huffman et al., 2001) was used to estimate the correlation of precipitation with EFs.  
390 This dataset is based on passive microwave measurements from the Special Sensor  
391 Microwave Imager (SSM/I), and infrared retrievals from the Geostationary Operational  
392 Environmental Satellite (GOES) and the Television InfraRed Observation Satellite (TIROS)  
393 Operational Vertical Sounder (TOVS). The monthly rainfall totals are corrected over some  
394 continental areas to match sparse ground-based observations, and at finer time scales the  
395 product relies exclusively on satellite-based precipitation estimates. We averaged the daily  
396 values to calculate a monthly average (mm/month) for the years 1997-2008, the period of  
397 availability. For EF measurements conducted before the year 1997, we used the monthly  
398  $2.5^{\circ} \times 2.5^{\circ}$  GPCPv2.1 precipitation product (Adler et al., 2003), which is available from 1979  
399 till present. Monthly averaged precipitation data for the years 1997-2008 were also used to  
400 define the mean annual precipitation (MAP). All data was regridded to  $0.5^{\circ} \times 0.5^{\circ}$  resolution  
401 using linear interpolation. Since we explored large-scale relations between EFs and the  
402 monthly and mean annual precipitation only, we may miss variability related to synoptic scale  
403 precipitation.

404 Temperature data were derived from a climatology and an anomaly source. The  
405 climatological data were downloaded from the Climate Research Unit (CRU) website  
406 (<http://www.cru.uea.ac.uk/>). We used the CRU CL 1.0 Mean Monthly Climatology product,  
407 with a resolution of  $0.5^{\circ} \times 0.5^{\circ}$  (New et al., 1999). This dataset gives the mean monthly surface

408 climate over global land areas, excluding Antarctica, and was interpolated from station data to  
409  $0.5^{\circ} \times 0.5^{\circ}$  for several variables. We then used the NASA GISS Surface Temperature Analysis  
410 (GISTEMP) as a source of temperature anomalies (Hansen et al., 1999). GISTEMP provides a  
411 measure of the global surface temperature anomaly with monthly resolution for the period  
412 since 1880, when a reasonable global distribution of meteorological stations was established.  
413 Input data for the analysis, collected by many national meteorological services around the  
414 world, is the unadjusted data of the Global Historical Climatology Network (Peterson and  
415 Vose, 1997). Documentation of the GISTEMP analysis is provided by Hansen et al. (1999),  
416 with several modifications described by Hansen et al. (2001). We used the 1961-1990  
417 anomalies with a 1200 km smoothing radius, which were downloaded from the NASA  
418 website (<http://data.giss.nasa.gov/gistemp/maps/>). The CRU climatology and GISTEMP  
419 anomalies were combined to estimate the monthly temperatures for the years 1967-2009.  
420 Monthly averaged temperature data for the years 1997-2008 were used to define the mean  
421 annual temperature (MAT).

422 The Normalized Difference Vegetation Index (NDVI) represents the amount of live green  
423 vegetation and its productivity, and may be a useful indication of vegetation characteristics  
424 (fuel abundance and also live fuel moisture conditions). Monthly Global Inventory Modelling  
425 and Mapping Studies (GIMMS) NDVI data with a  $8 \times 8$  km resolution (Tucker et al., 2005)  
426 were downloaded from the International Satellite Land Surface Climatology Project website  
427 (<http://islscp2.sesda.com/>). Different satellite series of NOAA's Advanced Very High  
428 Resolution Radiometer (AVHRR) were used for this NDVI record. The dataset consists of bi-  
429 monthly NDVI data for the years 1981 to 2006, which we averaged to monthly values. For EF  
430 measurements that were conducted before 1981 or after 2006, we used the monthly mean of  
431 the years 1981-2006.

432 The length of the dry season for the EF measurement locations was defined by counting the  
433 number of consecutive months in the 6-month period before the measurement was conducted  
434 with precipitation rates below 100 mm/month (GPCP  $1^{\circ} \times 1^{\circ}$  for the 1997-2008 period, and  
435 GPCPv2.1  $2.5^{\circ} \times 2.5^{\circ}$  for 1979-1997). This parameter partly overlaps with the precipitation  
436 rates, but the added value lies in containing a memory of precipitation; it may be an indicator  
437 of the precipitation conditions before the month of the actual measurement. It may be  
438 especially valuable for estimating the moisture content of fuels with low surface to volume  
439 ratios such as stems, which often take more than one month to come in equilibrium with

440 ambient moisture conditions (Bradshaw et al., 1984).

441

442

### 443 **3.4 Correlations between remotely sensed environmental data and EFs**

444 In Table 1 the correlation coefficients between the environmental data and the EFs of CO,  
445 CH<sub>4</sub>, CO<sub>2</sub>, and MCE (based on the EFs of CO and CO<sub>2</sub>) are given. Here, we lumped all the EF  
446 data of A&M2001-2009 for the three different biomes together. We performed simple linear  
447 regressions, with the EF as the dependant variable, and the different parameters that may  
448 control the EFs variability represent the independent variables. Besides the correlation  
449 coefficients (r), F-values were calculated to test if the regression between the EF and the  
450 different driver data was significant (if the F-value exceeds the critical value of  $F_{crit}$ , it  
451 indicates a significant fit). We also performed a multivariate regression to construct a  
452 regression equation that combined the different parameters that accounted for most of the EF  
453 variability, in order to see if different variables combined perform better than the variables  
454 separately, and to be able to construct EFs for grid cells where no measurements were  
455 performed.

456 For CO EF we found the highest correlation with FTC ( $r=0.49$ ) and NDVI ( $r=0.41$ ). The  
457 corresponding F-values (66.2 & 7.0) exceeded the critical F value ( $F_{crit}=6.7$ ) for a  
458 significance level of 0.01. When combining the different parameters in one regression  
459 equation, the correlation coefficient improved to 0.57. For the CH<sub>4</sub> EF, FTC ( $r=0.58$ ) and  
460 monthly precipitation ( $r=0.53$ ) were the most dominant parameters, and both correlations  
461 were significant at a level of 0.01. Using the additional information of each parameter  
462 increased the correlation ( $r=0.62$ ). For CO<sub>2</sub>, FTC and monthly precipitation yielded the  
463 highest descriptive power ( $r=-0.26$  and  $r=-0.37$ ), similar to CH<sub>4</sub>. Despite the relatively low  
464 correlation coefficients, both fits were significant with F-values of 10.1 and 27.1. The  
465 multivariate regression equation gave a slightly higher correlation ( $r=0.43$ ). In general, the  
466 highest correlations were found for FTC, which is not surprising since and this parameter  
467 covers the range from open grasslands, through savanna and woodlands, through tropical  
468 forest. Also, within biomes, FTC could explain part of the EF variability.

469 For MCE we found the highest correlation with monthly precipitation ( $r=-0.52$ ) and FTC ( $r=-$   
470 0.47), and both corresponding F-values (62.2 & 46.9) exceeded the critical F value for a

471 significance level of 0.01. All environmental parameters combined, the correlation coefficient  
472 improved to 0.58. For MCE we performed a similar analysis using the dataset of Akagi et al.  
473 (2010), which is based on EF data measured in fresh plumes only, which have not undergone  
474 significant photochemical processing. Overall, the correlations with the different  
475 environmental parameters did not improve compared to the EF dataset of A&M2001-2009; a  
476 maximum correlation coefficient of 0.55 was found using all environmental data combined.  
477 This is not an indication that one dataset is preferred above the other one; for CO and CO<sub>2</sub> it  
478 does not matter whether fresh or aged smoke is sampled. The differences could be the result  
479 of a larger number of samples in the A&M2001-2009 dataset. When translating our findings  
480 on MCE to other trace gases or aerosols, it may be preferable to use the Akagi et al. (2010)  
481 dataset because it consistently only takes those measurements focusing on fresh smoke into  
482 account, better representing initial emissions.

483 In general, repeating the calculations but focusing on each individual biome yielded lower  
484 correlations than with all measurements lumped together. However, some of the relations  
485 found when using the full suite of data were still valid. For example, also within the savanna  
486 and grassland biome we found a negative correlation between FTC and MCE (or positive  
487 correlation between FTC and the CO emission factor) with an almost identical slope and  
488 offset as when using all measurements. Correlations between the EFs and the environmental  
489 data for the extratropical forest were very poor. Possible explanations for these poor  
490 correlations are discussed in section 4. Higher correlations between EFs and the driving  
491 variables were found when focusing on specific locations, although it must be noted that the  
492 sample size of these correlations is relatively small. Figure 4a, 4b, and 4c show correlations  
493 for respectively Brazilian deforestation fires and savanna fires in Australia (FTC vs. MCE),  
494 Brazilian deforestation fires (FTC vs. CH<sub>4</sub> EF), and boreal fires in Alaska (precipitation vs.  
495 CH<sub>4</sub> EF). A similar pattern occurred when focusing on vegetation types: correlations between  
496 MCE and CH<sub>4</sub> EF were relatively low when using all data lumped together (Figure 2), and  
497 higher correlations were found in different individual studies, using a smaller sample size.  
498 Also, the extratropical forest data showed overall lower correlations than data for the savanna  
499 and tropical forest biome.

500

### 501 **3.5 Weighted EF averages**

502 Most large-scale biomass burning emission estimates are based on some combination of  
503 biomass or C combusted and EFs. These EFs are usually based on the arithmetic mean of a  
504 large number of measurements, most often using the work of A&M2001-2009. It is not  
505 known, however, whether the measurements are representative of the whole biome.  
506 Regionally, there is substantial variation in the density of measurements. For example, nearly  
507 all tropical forest measurements are made in the Brazilian Amazon and Yucatan province of  
508 Mexico (Figure 1), while information from other deforestation hot spots such as Bolivia and  
509 Indonesia is lacking. Different regional deforestation practices could in principle lead to  
510 variations in EFs, something that cannot be taken into account at the moment due to a lack of  
511 measurements. The same holds for the boreal region; according to the estimates of van der  
512 Werf et al. (2010), total C emissions from boreal Asia were almost 2.5 times as high as those  
513 from boreal North America in the last decade. Nevertheless nearly all the extratropical forest  
514 EF measurements were made in North America, and only one was conducted in boreal Asia  
515 (Figure 1).

516 While there are regional discrepancies in measurement locations, the measurements do  
517 capture most of the climate window in which most fires occur (Figure 5). To construct new  
518 weighted average EFs, we weighted each measurement with its quantitative importance in the  
519 fire-climate window. The size of the climatic window bins we used were 1° Celsius for mean  
520 annual temperature (MAT), 100 mm / year for mean annual precipitation (MAP), and 2% for  
521 fraction tree cover (FTC). Table 2 gives an overview of these new calculated mean values per  
522 biome. The weighted values are at most 18% different from the arithmetic mean, but mostly  
523 lower (Table 2). Some differences, however, can be noticed: EFs of CO were 8% below and  
524 13% above the mean of A&M2001-2009 for tropical forest and extratropical forest  
525 measurements, respectively. EFs of CH<sub>4</sub> were lower for each biome (16% on average). CO<sub>2</sub>  
526 EFs were somewhat lower for savannas (1.5%) and more variable for the tropical and  
527 extratropical biome.

528 On average, the weighted MCE for the different climate-windows was 7% and 11% higher  
529 than the A&M2001-2009 average for the tropical forest and extratropical biome, respectively.  
530 Weighted MCEs for the savanna biome were comparable with the biome-averaged values of  
531 A&M2001-2009. Overall, our new calculated weighted averages for CO, CH<sub>4</sub>, CO<sub>2</sub> EFs and  
532 MCE do not deviate much from the arithmetic mean of A&M2001-2009, and are well within  
533 the range of uncertainty, especially when also taking the substantial uncertainties in the GFED

534 fuel consumption estimates into account. This indicates that the measurement locations were  
535 representative with regard to emissions strengths. However, it does not provide information of  
536 the representativeness of the measurement locations for the whole biome, which will be  
537 addressed next.

538

539

### 540 **3.6 From a discrete towards a continuous classification scheme for EFs**

541 Following the work of Hoffa et al. (2003) and Ito and Penner (2005), we developed a non-  
542 vegetative classification scheme for EFs, driven by various environmental parameters. We  
543 performed a multivariate regression to construct an equation that combined the different  
544 environmental parameters (Table 1) for the CO, CH<sub>4</sub>, CO<sub>2</sub> EFs, and the MCE, which is a  
545 useful parameter since it can be related to many carboneous gases and certain aerosols (e.g.,  
546 Ward et al., 1996; Sinha et al., 2003; Yokelson et al., 2003). In Table 2 these new calculated  
547 mean values, weighted by the amount of biomass combusted in the 1997-2008 period, are  
548 given per biome. EFs of CO and CH<sub>4</sub> were ~13% and ~22% higher than the biome-averaged  
549 values of A&M2001-2009 for the savanna biome, and significantly lower for the tropical  
550 forest and extratropical forest biome. CO<sub>2</sub> EFs were the same for the savanna and tropical  
551 forest biome, and ~5% higher for the extratropical forest. The biome-averaged MCE deviates  
552 0.32%, 2.2%, and 1.2% from the biome-averaged values of A&M2001-2009 for savanna,  
553 tropical forest, and extratropical forest respectively.

554 Using the multivariate regression equation for MCE, which is mostly driven by monthly  
555 precipitation and FTC (Table 1), we constructed monthly MCE fields with a spatial resolution  
556 of 0.5°×0.5° for the years 1997-2008. In Figure 6a the newly calculated MCE, weighted by  
557 the amount of biomass combusted in the 1997-2008 period, is shown on a global scale. In  
558 general, tropical forest and boreal areas show lower MCE values compared to savanna  
559 regions. Spatial differences within savanna areas are obvious as well; woodland areas (for  
560 example, in Angola) have a relatively low MCE compared to areas where grasslands or open  
561 savannas are the dominant vegetation type, for example in South Africa or in the Australian  
562 interior.

563 In Figure 6b the difference between our new “continuous“ MCE and the biome-dependent  
564 MCE of A&M2001-2009 is shown. The latter was constructed using the MOD12Q1 land

565 cover map for 2001 (Friedl et al., 2002) to distribute the biome-specific MCEs over the globe.  
566 Areas where we predict a lower MCE, and thus emit relatively more reduced gases (CO,  
567 CH<sub>4</sub>), are shown in blue. We expect that these grid cell specific MCEs are more reliable in the  
568 tropics than in boreal regions because more measurement locations were in the tropics. This  
569 may also be why FTC and monthly precipitation were the two most important parameters. In  
570 addition, the regression cannot deal with agricultural waste burning and peat burning regions,  
571 and these regions will receive biome-specific EFs. Regarding the savanna and grassland  
572 biome: we found the highest MCE in Australia (0.9466), followed by southern hemisphere  
573 Africa (0.9432), northern hemisphere South America (0.9403), and southern hemisphere  
574 South America (0.9386). Although differences in MCE are relatively small, they have a  
575 substantial influence on the amount of CO and other reduced trace gases released. For  
576 example, the small difference in MCE between Australia and southern hemisphere South  
577 America (~0.9%) may imply a relatively large difference in the amount of CO emitted  
578 (~16%) if the total amount of C emitted as CO and CO<sub>2</sub> is kept constant in both regions. An  
579 important next step is to implement these spatial and temporal EF and MCE scenarios into  
580 GFED, and quantify regional differences in trace gasses emitted.

581

## 582 **4 Discussion**

583 We evaluated a large body of available literature describing EF measurements conducted in  
584 different biomes throughout the world, and explored the relations between the EFs and global  
585 low-resolution datasets of parameters that may influence EF variability. We chose to compare  
586 EFs with seven important control parameters for which global datasets were available and  
587 extended back to at least the early 1990s. These could account for up to about 32.5% ( $r=0.57$ ),  
588 38.4% ( $r=0.62$ ), 18.5% ( $r=0.43$ ), and 33.6% ( $r=0.58$ ) of the variability for respectively CO,  
589 CH<sub>4</sub>, CO<sub>2</sub>, and MCE. Several factors may account for the remaining variability and are  
590 discussed in section 4.1 - 4.4. We discuss the new weighted biome-averaged EFs in section  
591 4.5, followed by recommendations for new EF campaigns (section 4.6) and our future steps  
592 (section 4.7).

593

#### 594 **4.1 Uncertainty in environmental parameters**

595 Monthly averages of coarse-resolution (regridded to  $0.5^{\circ}\times 0.5$ ) data were used to assess fire  
596 emissions, fraction tree cover, precipitation, temperature, NDVI, and the length of the dry  
597 season for the different EF measurement locations. The use of spatially and temporal higher  
598 resolution data is preferred over lower resolution data, but detailed information on the  
599 location and date of the measurements was often lacking. Even if detailed information was  
600 given, a large number of EF measurements were conducted in the 1980s and early 1990s, for  
601 which period global datasets are often lacking at sufficient high resolution. Also in more  
602 recent periods data availability would limit more detailed analyses: while FTC is available at  
603 500-meter resolution, it is only available for the year 2002. And since fires likely impact FTC  
604 a multi-year product is required for consistency, so that –for example- each EF measurement  
605 can be linked to the FTC before the fire. Here we have not included uncertainties in these  
606 environmental parameters because they have not undergone an official error assessment, with  
607 the exception of the precipitation data.

608

#### 609 **4.2 Additional drivers of emission factor variability**

610 Although other environmental data (e.g. precipitation duration, fuel spacing, wind, and  
611 topography) may play an important role in fire characteristics and thus in the partitioning of  
612 trace gases emitted (e.g. Lobert et al., 1991), we could not take these factors into account  
613 because reliable information is not available from global datasets (see Section 4.1). Only few  
614 papers describing the measurements include detailed information on climatic and  
615 environmental conditions. Fuel composition may be another crucial factor for EF partitioning  
616 that was not taken into account here, and which may account for part of the variability not  
617 captured by the 7 parameters we could include because consistent information was available  
618 for all measurement locations.

619 In the future, a combination of 1) more EF field measurements, 2) better use of simultaneous  
620 satellite retrievals of trace gases (e.g., CO and NO<sub>2</sub>), and 3) the availability of higher spatial  
621 and temporal resolution satellite datasets may further improve our understanding of how  
622 certain environmental parameters influence the EF variability for specific fires.

623

### 624 **4.3 Different measurement approaches and techniques**

625 Various analytical techniques have been used in recent field experiments, like non-dispersive  
626 infrared analysis (NDIR), Fourier transform infrared spectroscopy (FTIR), and gas  
627 chromatography. Detailed descriptions of these different techniques can be found in the  
628 literature (Ward and Radke, 1993; Yokelson et al., 1999; Christian et al., 2004). For real-time  
629 concentration measurements, the analytical instruments must be close to the fire. A distinction  
630 can be made between ground-based (tower, mast) and airborne (airplane, helicopter)  
631 measurements. Airborne measurements sample an integrated mixture of the emissions from  
632 both combustion types (smoldering and flaming). For ground-based measurements, which  
633 have a smaller footprint, the separation between smoldering and flaming combustion is more  
634 clear, but even here both processes occur simultaneously in a given patch at most times.  
635 Ground-based sampling probably oversamples the emissions which tend to be emitted during  
636 less vigorous phases of a fire and therefore remain closer to the ground, while airborne  
637 sampling may be biased towards emissions from the flaming phase that rise to higher altitudes  
638 (Andreae et al., 1996; Yokelson et al., 2008). Airborne measurements of chaparral  
639 vegetation in California (Laursen et al., 1992) were for example compared to ground-based  
640 measurements of the same vegetation type (Ward and Hardy, 1989), with overall lower EFs  
641 for CO (18%) and CH<sub>4</sub> (60%), and higher CO<sub>2</sub> (5%) due to the bias towards the flaming  
642 phase. Yokelson et al. (2008) performed a similar analysis for tropical forest fires, and also  
643 found lower EFs of CO and CH<sub>4</sub> for airborne measurements.

644 Although differences between measurement techniques are more important for sticky or  
645 reactive gases, the use of different techniques may have caused variations in CO, CH<sub>4</sub>, and  
646 CO<sub>2</sub> EFs measured in specific experiments. For example, SAFARI campaign measurements  
647 were conducted in South Africa and Zambia, and different research groups were involved to  
648 estimate EFs. Airborne Fourier transform infrared spectroscopy (AFTIR) was used by  
649 Yokelson et al. (2003) to measure EFs, while Sinha et al. (2003) used gas chromatography.  
650 Both measuring techniques gave different EFs of CO, CH<sub>4</sub> and CO<sub>2</sub>, even though the location  
651 and timing of the burning event was identical. Another example comes from extratropical  
652 forest biome; the use of different analytical techniques led to a difference of 23% for CO, 8%  
653 for CH<sub>4</sub>, and 2% for CO<sub>2</sub> EFs for the same fires in North America (Hegg et al., 1990; Laursen  
654 et al., 1992).

655

#### 656 **4.4 Flaming/smoldering assumptions**

657 The ratio between flaming and smoldering combustion of a fire is crucial for estimating the  
658 overall EF for different trace gases. In savanna fires, for example, flaming combustion  
659 dominates, and the EF for reduced species is relatively low compared to forest fires where the  
660 smoldering phase is often more important. The proportion of flaming and smoldering  
661 combustion can vary considerably also within fires in the same biome as a function of internal  
662 parameters (for example moisture content). It may seem desirable to provide separate EFs for  
663 flaming and smoldering combustion, but this is not always possible given the data available.  
664 In the field, EFs are generally determined by averaging several instantaneous measurements  
665 from the fire. Most emissions are assumed to be a mixture of flaming and smoldering  
666 combustion, and it is essential that averaging of both phases is done correctly when the EF for  
667 an entire fire is sought. Generally the individual measurements are weighted according to the  
668 amount of fuel combusted in the time interval represented by the measurement (Ward and  
669 Hardy, 1991). This approach requires information that is only available in experimental fires  
670 in the laboratory or to a limited extent in the field, so often assumptions had to be made on the  
671 flaming to smoldering ratio leading to another source of uncertainty and potential to yield  
672 different EFs for similar smoke plumes.

673 Estimates of the relative importance of the flaming and smoldering phases vary in literature;  
674 for grass and shrub fires flaming combustion dominates and likely accounts for 80% to 90%  
675 of fuel consumption (Shea et al., 1996; Ward et al., 1996). For tropical forest and boreal fires  
676 smoldering combustion is more important. Bertschi et al. (2003), for example, assumed that  
677 the smoldering and flaming phases combusted equal amounts of biomass in boreal areas, and  
678 residual smoldering measurements were combined with airborne measurements of Goode et  
679 al. (2000) to calculate an overall EF. For African miombo fires a flaming-smoldering ratio of  
680 90-10 was taken, and airborne FTIR measurements from a study of Yokelson et al. (2003)  
681 were used to represent the flaming part. A change in these flaming-smoldering ratio's will  
682 impact the overall EF substantially, so the assumptions made by different authors are  
683 therefore important to consider (Yokelson et al., 1996).

684 A&M2001-2009 made the assumption that when smoldering and flaming emissions were  
685 given separately in ground-based studies, the emissions were combined to represent the  
686 complete fire. For this purpose A&M2001-2009 either used data on the fractions of fuel  
687 combusted in the smoldering and flaming stages provided in a given study, or, when this

688 information was not available, typical values from other studies on the same type of fire were  
689 used.

690

#### 691 **4.5 Weighted means**

692 The biome-averaged EF values of A&M2001-2009 are widely used in the modeling  
693 community. These mean values may not be representative for the whole biome (e.g. nearly all  
694 extratropical forest measurements were made in North America, and only one measurement  
695 was made in Siberia). We performed two levels of weighting. First, by placing the  
696 measurements in their climatic window (based on mean annual precipitation, mean annual  
697 temperature, and fraction tree cover) we were able to weigh the different measurements with  
698 regard to their quantitative importance, using the GFED3.1 C emissions estimates in the  
699 corresponding C climatic window. The weighted EFs are within 6.7%, 7.9%, and 13.2% of  
700 the arithmetic mean of A&M2001-2009 for CO, 17.4%, 15.2%, and 6.7% for CH<sub>4</sub>, and 2.1%,  
701 7.2%, and 11.4% for the MCE for the savanna, tropical forest, and extratropical forest biome,  
702 respectively. The weighted EFs of CO<sub>2</sub> are within 3% of the arithmetic mean for all three  
703 biomes. According to the linear regression results for the different EF drivers, the climatic  
704 window with the most predictive power for CO, CH<sub>4</sub> and CO<sub>2</sub> EFs together is based on  
705 fraction tree cover and mean annual precipitation (Table 2, FTC-MAP). Based on the  
706 weighting by FTC and MAP, the EFs are systematically lower than the arithmetic mean of  
707 A&M2001, with a 8.7%, 3.7%, and 2.1% decrease for CH<sub>4</sub>, and 1.2%, 1.5%, and 0.4% for  
708 CO<sub>2</sub>, for the savanna, tropical forest, and extratropical forest biome, respectively. For CO the  
709 weighted EFs were lower than the arithmetic mean of A&M2001-2009 for savanna and  
710 tropical forest (1.7% and 7.9%), but higher for extratropical forest (3.8%).

711 We adjusted the different vegetation types that were defined by A&M2001-2009, and based  
712 on these biomes (savanna and grasslands, tropical forest, extratropical forest), we calculated  
713 new weighted EF averages. Specifically, several measurements were conducted in vegetation  
714 types (for example chaparral in California and pinetree forest in Mexico) that cannot be  
715 clearly classified as savanna and grassland, tropical forest, or extratropical forest. While the  
716 savanna and tropical forest biome EF measurements were clustered in Figure 5, the  
717 extratropical forest measurements show more variation (Figure 5b). For a more specific EF  
718 average, it could be helpful to expand the amount of vegetation types, for example by adding  
719 a 'temperate forest' and/or 'chaparral' biome as in the Akagi et al. (2010) database.

720 Second, another level of weighting was performed by moving from a discrete classification  
721 based on a limited number of biome types, to stratifying EFs by vegetation density (FTC,  
722 NDVI) and climatic conditions (precipitation, temperature, length of dry season). Therefore,  
723 we developed a non-vegetative classification scheme for EFs (Figure 6), driven by the  
724 different environmental parameters presented in Table 1. The global average MCE, weighted  
725 by the amount of biomass combusted in the 1997-2008 period, for the whole savanna biome  
726 compared well with the biome-averaged MCEs of A&M2001-2009 and the weighted average  
727 MCEs for the different climate windows (Table 2). However, CO and CH<sub>4</sub> EFs were ~13%  
728 and ~22% higher than the biome-averaged values of A&M2001-2009 for the savanna biome,  
729 possibly linked to the underrepresentation of woodland fires in EF measurements. In addition,  
730 regional differences in MCE for the savanna biome were found, with the highest MCE for  
731 savanna & grasslands in Australia. Although our temporal and spatial variable MCE captures  
732 the grassland to closed savanna range in the savanna biome reasonably well, future  
733 adjustments in our scheme are needed –for example for extratropical forests- because it may  
734 be biased towards tropical regions where the majority of measurements were made.

735

#### 736 **4.6 Recommendations for future EF campaigns**

737 Ongoing studies aim to better quantify EFs. They often fill a niche, for example by measuring  
738 fuels for which information is lacking, like tropical peat fires. In addition, emphasis has  
739 switched towards understanding chemical processes within the fire plume. We have shown,  
740 however, that current available information on EFs is insufficient to improve our  
741 understanding of the factors driving variability in EFs to levels of uncertainty found in other  
742 fire emissions parameters. By taking into account the following recommendations this  
743 situation may be improved:

744 *Spatial representation:* several areas are undersampled but are key emissions areas, most  
745 importantly Central Africa, boreal Asia, and Indonesia. Each of these regions likely has  
746 relatively high rates of emissions of reduced gases; more woodland burning in Central Africa  
747 compared to southern Africa where most savanna measurements were made, more  
748 groundfires in boreal Asia compared to boreal North America where most extratropical EFs  
749 were measured, and moister conditions and more peat burning in Indonesia compared to  
750 South America where most deforestation fire EFs were made.

751 *Seasonality*: to better understand the temporal variation of EFs in specific vegetation biomes,  
752 there is a need of measurements made over the full fire season, following Korontzi et al.  
753 (2003). In addition, the currently available measurements have placed too much weight to the  
754 months before (tropical forest) or the months before and after (grassland and savannas) the  
755 peak fire month and a stronger focus towards the peak fire month would yield a better sample  
756 of the fire seasonality.

757 *Fuel and ambient conditions*: measuring and describing fuel composition, its moisture  
758 content, and ambient conditions such as windspeed and temperature may allow for a better  
759 understanding of the factors driving EFs, especially when multiple locations are visited with  
760 the same measurement protocol. This requires a more multi-disciplinary approach and calls  
761 for combining campaigns aiming to quantify biomass loads, combustion completeness, EFs,  
762 and satellite validation of e.g. hotspot detection efficiency and the accuracy of burned area.

763

#### 764 **4.7 Future steps**

765 We found that stratifying EFs by vegetation density (fraction tree cover) and climatic  
766 conditions may better represent the large variability in EFs compared to a discrete  
767 classification based on a limited number of biome types. Based on these findings we aim to  
768 implement different EF scenario's into the GFED modeling framework. In combination with  
769 inverse modeling and space-based observations of trace gases, we will then investigate  
770 whether these new estimates corresponds better with atmospheric constraints.

771

### 772 **5 Conclusion**

773 The partitioning of combusted biomass into trace gases and aerosols shows large variation in  
774 time and space. We assessed what fraction of this variability can be explained by coarse  
775 resolution, globally available datasets including fraction tree cover, precipitation, and  
776 temperature. When combined, these datasets could account for up to about 40% ( $r=0.62$ ) of  
777 the variability in emission factors. Uncertainties in driver data, the range of fuel C content,  
778 differences in measuring techniques, assumptions on weighting ratios of flaming and  
779 smoldering contributions, and insufficient information on the measurements may account for  
780 part of the remaining variability. In addition, we neglected driver data such as fuel spacing,  
781 topography, and windspeed, which also may explain part of the variability.

782 We have calculated new average EFs for three biomes, by 1) weighting the EF locations by  
783 the amount of biomass combusted, and 2) building new maps of MCE using the relations  
784 between environmental variables and EFs, and weigh each grid cell by the amount of biomass  
785 combusted. Using the climatic window with the highest predictive power, weighted EFs for  
786 the individual EF measurement locations were lower than the arithmetic mean of A&M2001-  
787 2009, with a 8.7%, 3.7%, and 2.1% decrease for CH<sub>4</sub>, and 1.2%, 1.5%, and 0.4% for CO<sub>2</sub>, for  
788 the savanna, tropical forest, and extratropical forest biome, respectively. For CO the weighted  
789 EFs were lower than the arithmetic mean of A&M2001-2009 for savanna and tropical forest  
790 (1.7% and 7.9%), and higher for extratropical forest (3.8%). Taking all levels of uncertainty  
791 into account, none of these differences may be significant.

792 However, the second level of weighting using a non-vegetative classification EF scheme  
793 driven by different environmental parameters indicated that the MCE for savanna and  
794 grasslands may be lower than the MCE based on the arithmetic mean of all EF measurements.  
795 This would indicate higher emissions of CO and other reduced gases for the same amount of  
796 biomass burned for all global grasslands and savannas combined due to an  
797 underrepresentation of EF measurements in woodland burning regions. In addition, regional  
798 differences in MCE for the savanna biome were found, with the highest MCE (and thus  
799 lowest CO EF) for savanna & grasslands in Australia.

800 Currently, most of the literature describing emission factor measurements lack a detailed  
801 description of the measurement site and ambient conditions during the experiment. This  
802 information is crucial to better understand the differences between the various measurements,  
803 and be able to understand the representativeness of large-scale satellite data and ambient  
804 conditions as done in this study. In addition, to better facilitate our understanding and ability  
805 to model MCE or EFs, more EF measurements should be performed in the peak fire months  
806 and in unsampled geographic areas. The development of a more uniform sampling protocol  
807 for the sampling and measurements of EFs in different vegetation types is another crucial step  
808 to better compare different measurements. For example, the database of Akagi et al. (2010),  
809 that compiles EFs based on a more uniform and accurate sampling protocol consistently only  
810 takes those measurements focusing on fresh smoke into account, better representing initial  
811 emissions.

812 A future step will be to implement our findings into the Global Fire Emission Database  
813 (GFED), and in combination with inverse modeling and space-based observations of trace

814 gases, to investigate how a better representation of the spatial and temporal variability in EFs  
815 may improve our understanding of biomass burning emissions.

816

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825

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1119 **Tables:**

1120 Table 1. Correlation coefficients (r) and F-values (F) for CO, CH<sub>4</sub>, and CO<sub>2</sub> EF measurements  
 1121 and different driver data. The MCE, based on the CO and CO<sub>2</sub> EF, are also shown. The  
 1122 correlation coefficient for the multivariate regression equation is also shown (r combined). *n*  
 1123 corresponds to the number of samples used, and F-values shown in italic indicate relations  
 1124 that did not exceed the critical F-value for a significance level of 0.01.

| Driver data               | CO (n=216)   |             | CH <sub>4</sub> (n=205) |              | CO <sub>2</sub> (n=169) |             | MCE (n=169)  |             |
|---------------------------|--------------|-------------|-------------------------|--------------|-------------------------|-------------|--------------|-------------|
|                           | r            | F           | r                       | F            | r                       | F           | r            | F           |
| Fraction tree Cover       | <b>0.49</b>  | <b>66.2</b> | <b>0.58</b>             | <b>104.3</b> | <b>-0.26</b>            | <b>10.1</b> | <b>-0.47</b> | <b>46.9</b> |
| Monthly Precipitation     | <b>0.40</b>  | <i>1.9</i>  | <b>0.53</b>             | <b>13.8</b>  | <b>-0.37</b>            | <b>27.1</b> | <b>-0.52</b> | <b>62.2</b> |
| Mean Annual Precipitation | <b>0.29</b>  | <b>3.2</b>  | <b>0.33</b>             | <b>4.4</b>   | <b>-0.13</b>            | <i>0.4</i>  | <b>-0.15</b> | <b>4.1</b>  |
| Monthly Temperature       | <b>-0.13</b> | <i>0.1</i>  | <b>0.03</b>             | <i>0.1</i>   | <b>-0.13</b>            | <b>2.7</b>  | <b>0.01</b>  | <i>0.2</i>  |
| Mean Annual Temperature   | <b>-0.23</b> | <i>1.1</i>  | <b>-0.24</b>            | <b>2.2</b>   | <b>0.16</b>             | <i>0.9</i>  | <b>0.29</b>  | <b>15.9</b> |
| Monthly NDVI              | <b>0.41</b>  | <b>7.0</b>  | <b>0.39</b>             | <i>0.5</i>   | <b>-0.22</b>            | <i>0.2</i>  | <b>-0.46</b> | <b>46.1</b> |
| Length dry season <100mm  | <b>0.17</b>  | <b>22.1</b> | <b>-0.06</b>            | <i>0.6</i>   | <b>0.03</b>             | <b>5.9</b>  | <b>-0.05</b> | <i>0.4</i>  |
| r combined                | <b>0.57</b>  |             | <b>0.62</b>             |              | <b>0.43</b>             |             | <b>0.58</b>  |             |

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1130 Table 2. EFs of CO, CH<sub>4</sub>, CO<sub>2</sub> (in g/kg DM), and MCE for savanna (S), tropical forest (T),  
 1131 and extratropical forest (E), weighted by carbon emissions and stratified by mean annual  
 1132 precipitation (MAP), mean annual temperature (MAT), fraction tree cover (FTC) bins, and a  
 1133 multivariate regression equation that combined different environmental parameters (Table 1).  
 1134 Biome-averaged arithmetic means of A&M2001-2009 are also shown, with standard  
 1135 deviations in parenthesis. The results for the climatic window with the highest predictive  
 1136 power are shown in *italic*.

|  | CO (g/kg DM)             |                           |                           | CH <sub>4</sub> (g/kg DM)  |                            |                            | CO <sub>2</sub> (g/kg DM)  |                            |                             | MCE          |              |              |
|--|--------------------------|---------------------------|---------------------------|----------------------------|----------------------------|----------------------------|----------------------------|----------------------------|-----------------------------|--------------|--------------|--------------|
|  | S                        | T                         | E                         | S                          | T                          | E                          | S                          | T                          | E                           | S            | T            | E            |
| Mean Annual<br>Precipitation – Mean<br>Annual Temperature      | <b>56</b>                | <b>94</b>                 | <b>107</b>                | <b>1.9</b>                 | <b>5.6</b>                 | <b>4.0</b>                 | <b>1624</b>                | <b>1636</b>                | <b>1588</b>                 | <b>0.948</b> | <b>0.919</b> | <b>0.911</b> |
| Fraction Tree Cover –<br>Mean Annual<br>Temperature            | <b>61</b>                | <b>97</b>                 | <b>120</b>                | <b>2.1</b>                 | <b>5.8</b>                 | <b>4.7</b>                 | <b>1622</b>                | <b>1615</b>                | <b>1529</b>                 | <b>0.944</b> | <b>0.915</b> | <b>0.911</b> |
| <i>Fraction Tree Cover –<br/>Mean Annual<br/>Precipitation</i> | <i>59</i>                | <i>93</i>                 | <i>112</i>                | <i>2.1</i>                 | <i>5.7</i>                 | <i>4.7</i>                 | <i>1627</i>                | <i>1578</i>                | <i>1565</i>                 | <i>0.949</i> | <i>0.917</i> | <i>0.911</i> |
| Environmental parameters<br>combined                           | <b>68</b>                | <b>82</b>                 | <b>95</b>                 | <b>2.8</b>                 | <b>4.6</b>                 | <b>4.2</b>                 | <b>1647</b>                | <b>1627</b>                | <b>1648</b>                 | <b>0.943</b> | <b>0.930</b> | <b>0.911</b> |
| A&M2001-2009   | <b>60</b><br><b>(19)</b> | <b>101</b><br><b>(16)</b> | <b>106</b><br><b>(36)</b> | <b>2.3</b><br><b>(0.8)</b> | <b>6.6</b><br><b>(1.8)</b> | <b>4.8</b><br><b>(1.8)</b> | <b>1646</b><br><b>(99)</b> | <b>1626</b><br><b>(39)</b> | <b>1572</b><br><b>(106)</b> | <b>0.946</b> | <b>0.911</b> | <b>0.911</b> |

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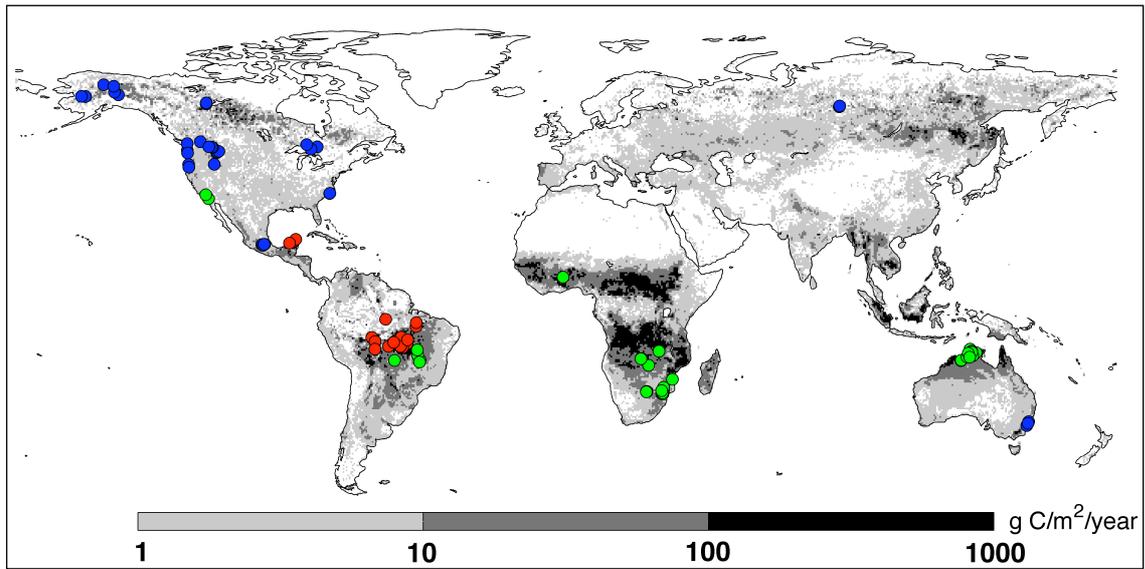
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1142 **Figures**



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1144 **Figure 1:** Locations where simultaneous CO and CO<sub>2</sub> EFs were measured. Locations were  
1145 stratified by biome following A&M2001; savanna & grassland (purple), tropical forest (red),  
1146 and extra-tropical forest (yellow). Background map shows annual GFED3.1 fire emissions in  
1147  $\text{g C/m}^2/\text{year}$ , averaged over 1997-2008, and plotted on a log scale.

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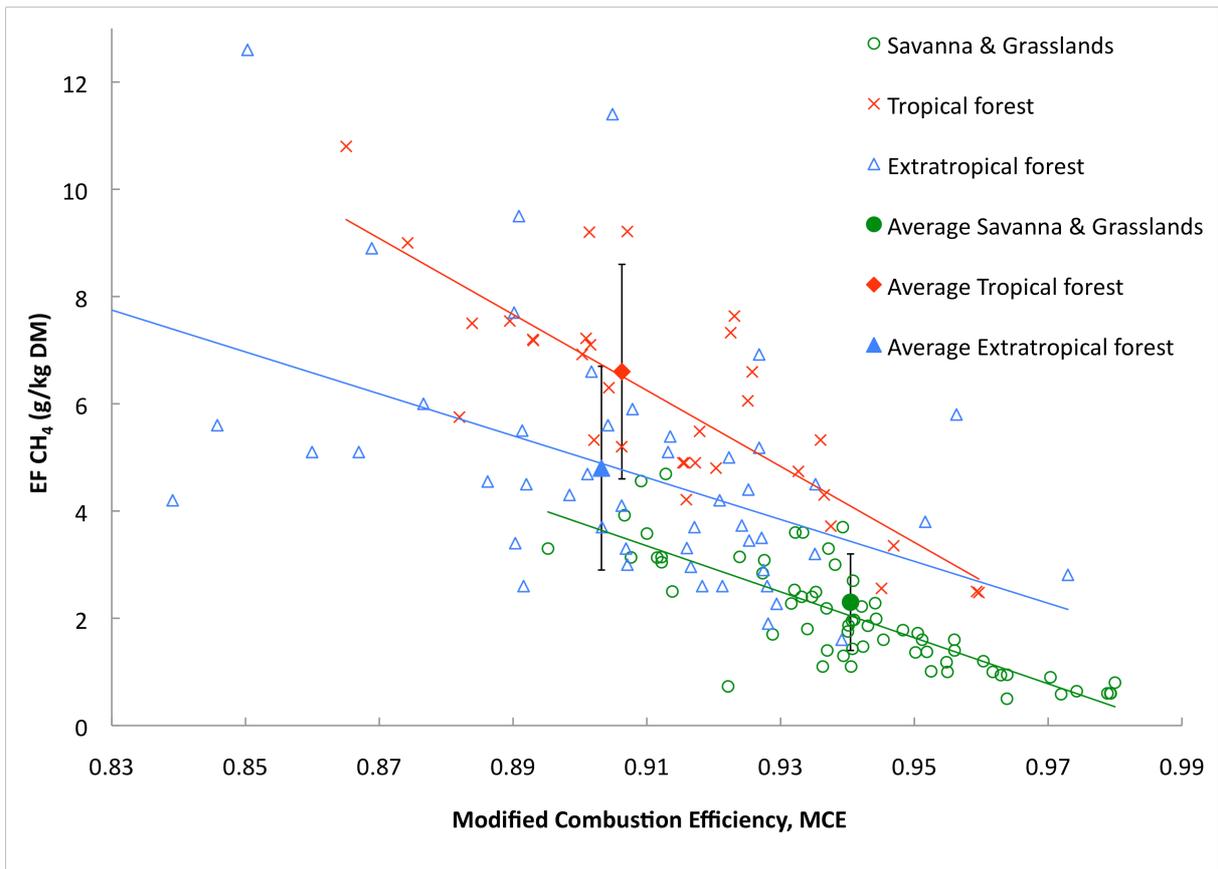
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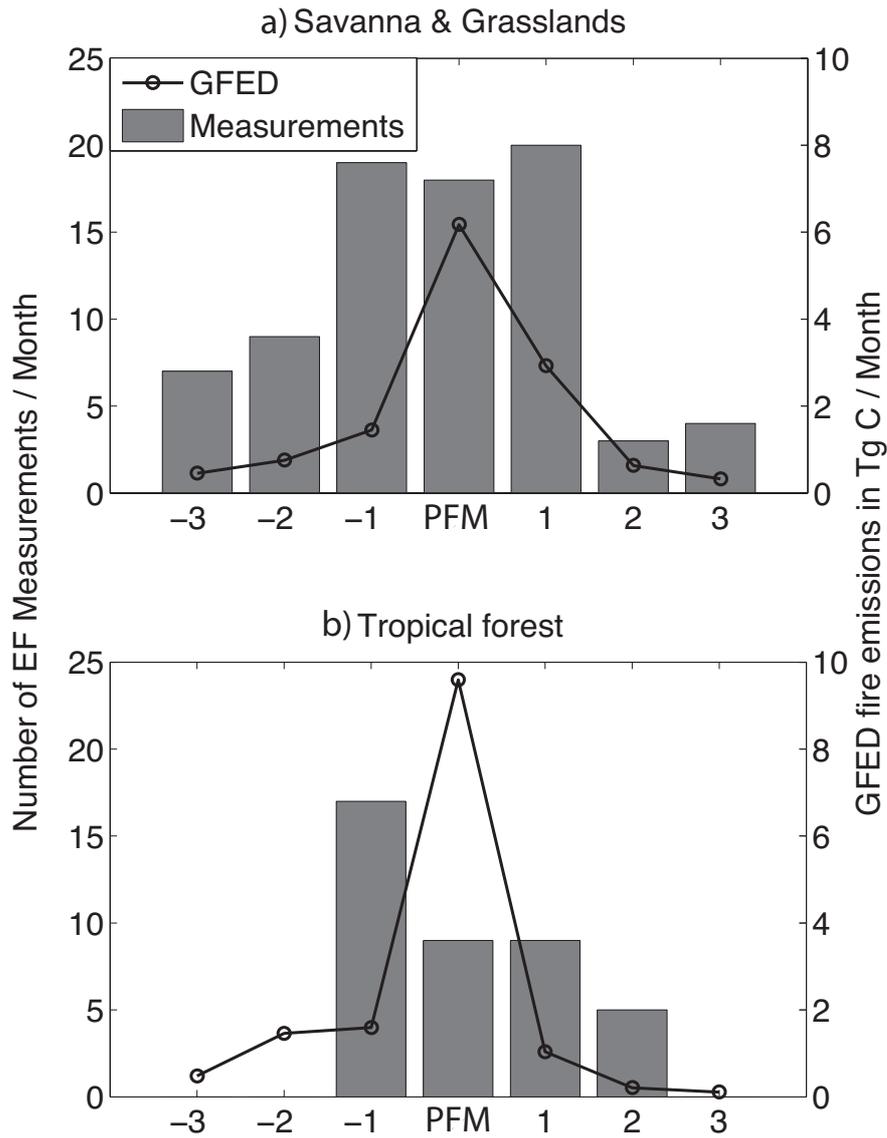
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1157 **Figure 2:** Methane (CH<sub>4</sub>) EFs and the molar-based modified combustion efficiency (MCE)  
 1158 for all available measurements, the biome-averaged values presented in A&M2001-2009, and  
 1159 regression lines. The errorbar indicates the standard deviation as reported in A&M2001-2009.  
 1160 Regression coefficients for the different biomes can be found in the text (Section 3.2, lines  
 1161 292-299).

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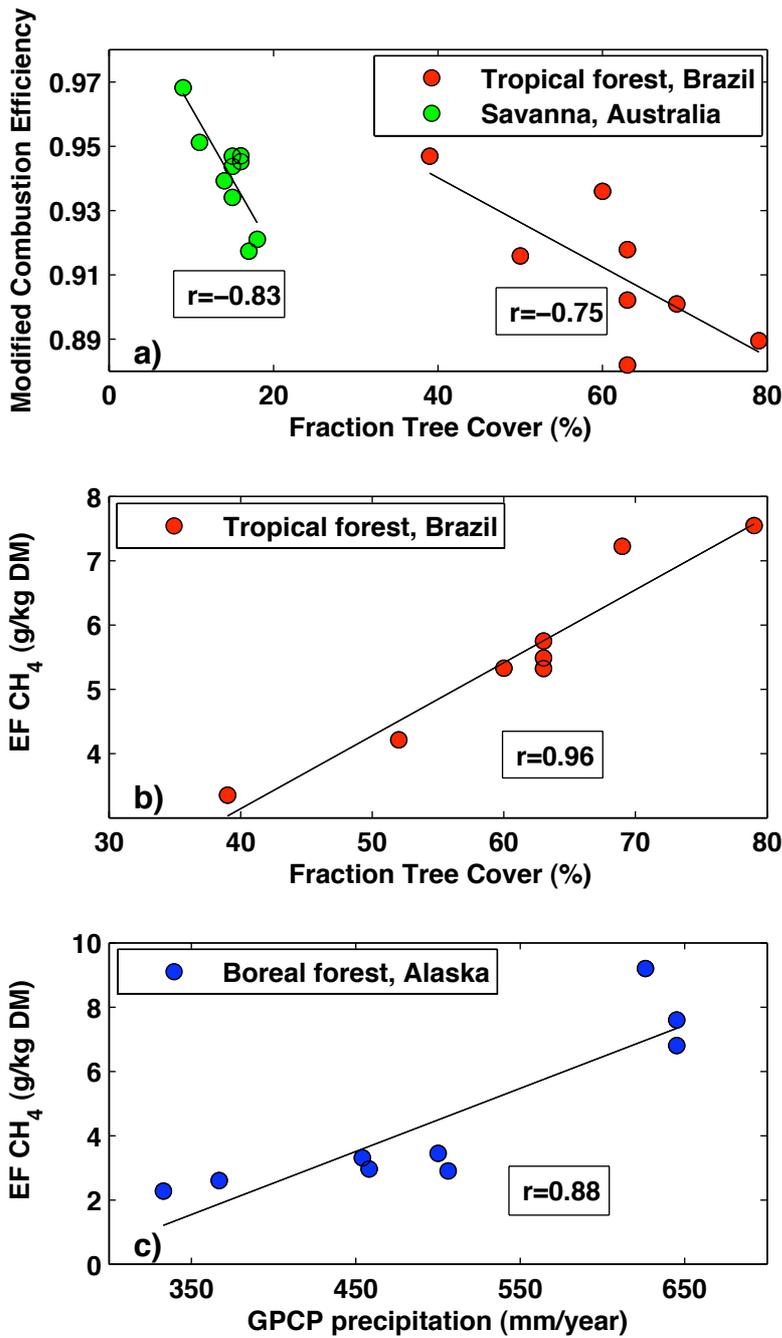


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1165 **Figure 3:** Number of EF measurements (bar) and GFED3.1 fire emissions (line) in Tg C for  
 1166 the peak fire month (PFM), and the months before and after the PFM, for all EF measurement  
 1167 locations in the a) savanna and grassland and b) tropical forest biome.

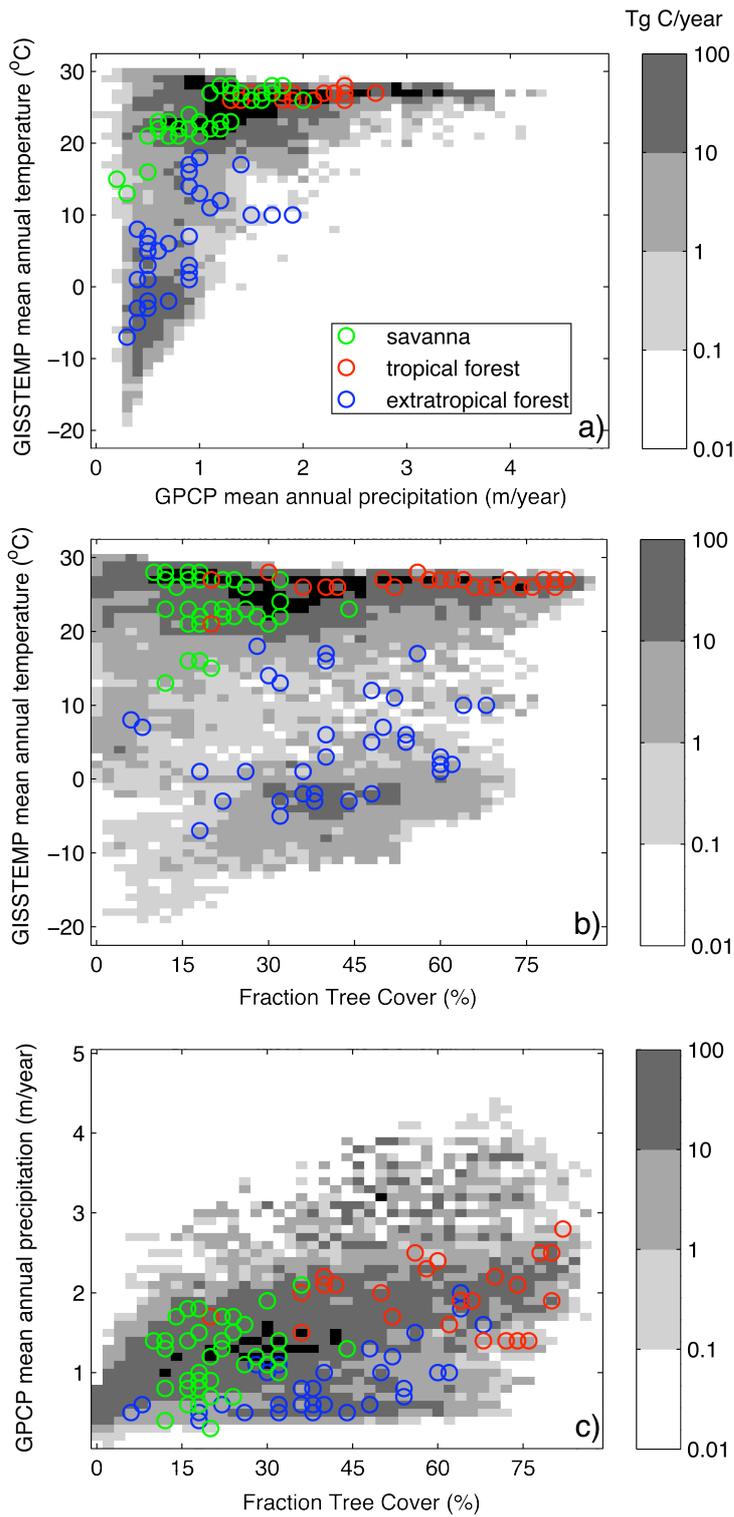
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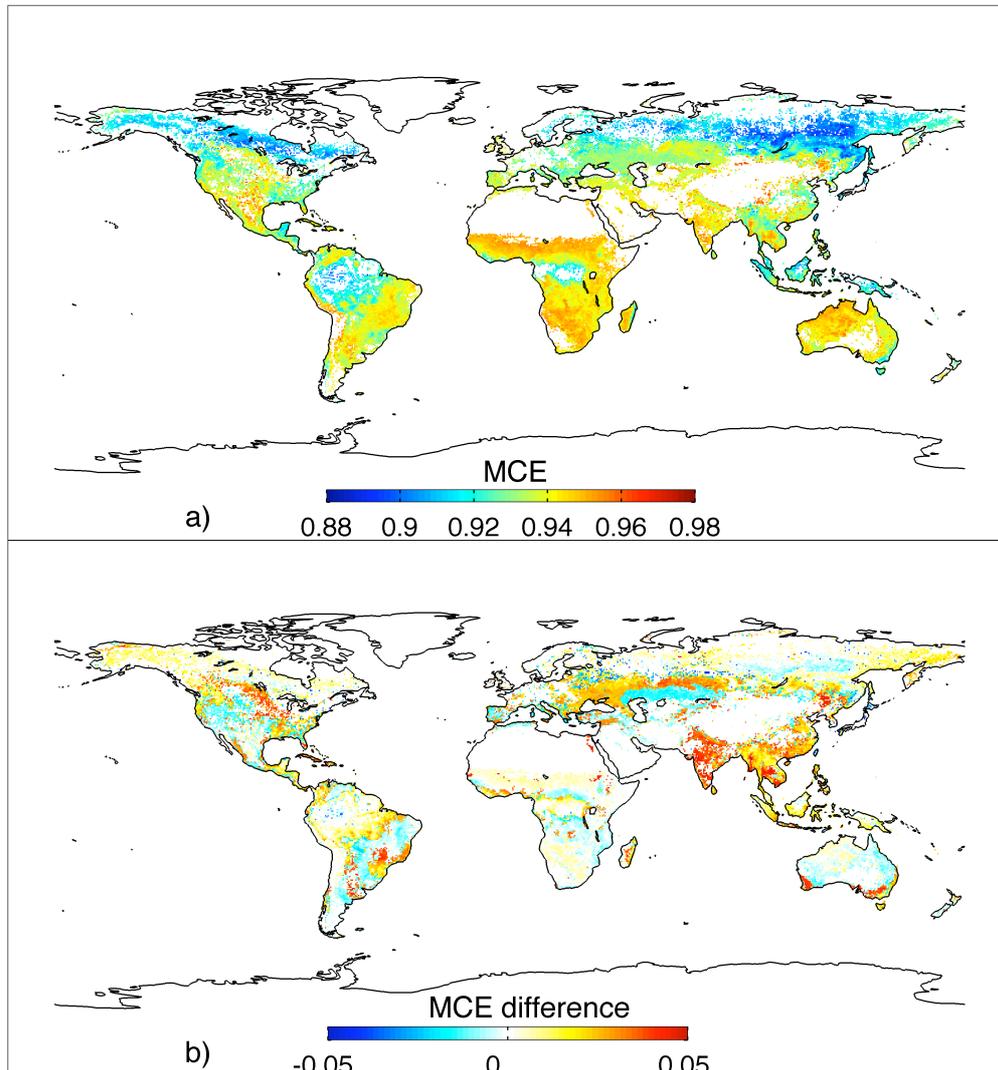
1170 **Figure 4:** Relations between driver data and EFs or MCE for selected regions. a) fraction tree  
 1171 cover and modified combustion efficiency (MCE) for savanna measurements in Australia  
 1172 (Hurst et al., 1994; Shirai et al., 2003) and tropical deforestation measurements in Brazil  
 1173 (Yokelson et al., 2007), b) fraction tree cover and CH<sub>4</sub> EF for tropical deforestation  
 1174 measurements in Brazil (Yokelson et al., 2007), and c) precipitation and CH<sub>4</sub> EF for extra-  
 1175 tropical forest measurements in Alaska (Laursen et al., 1992; Goode et al., 2000; Wofsy et al.,  
 1176 1992; Nance et al., 1993).



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1178 **Figure 5:** GFED3.1 fire emissions in Tg C/year (mean for 1997-2008) in a temperature –  
 1179 precipitation (a), temperature – fraction tree cover (b), and precipitation – fraction tree cover  
 1180 (c) window overlain by EF measurements in savanna and grasslands (green), tropical forest

1181 (red), and extratropical forest (blue circles). Temperature and precipitation were averaged over  
1182 1997 – 2008.



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1184 Figure 6. a) MCE based on a multivariate regression equation that combined different  
1185 environmental parameters (see Section 3.5), with a spatial resolution of  $0.5^{\circ} \times 0.5^{\circ}$  and  
1186 weighted by the amount of biomass combusted according to GFED3.1 for the years 1997-  
1187 2008. b) Difference between a) and the biome average MCE according to data of A&M2001-  
1188 2009. Here, emissions from peat fires have been neglected.

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