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

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Fires are a major source of trace gases and aerosols to the atmosphere. Quantitative knowledge on biomass burned is improving, most importantly due to new burned area datasets. The partitioning of biomass burned into emitted trace gases and aerosols, however, has received relatively little attention. To convert estimates of biomass burned to trace gas and aerosol emissions, most studies have used emission ratios (or emission factors (EFs)) based on the arithmetic mean of field measurement outcomes, stratified by biome. However, EFs vary substantially in time and space, even within a single biome. In addition, it is unknown whether the measurement locations provide a representative sample for the various biomes. Here we used the available body of EF literature in combination with satellite-derived information on vegetation characteristics and climatic conditions to better understand the spatio-temporal variability in EFs. While focusing on CO, CH₄, and CO₂, our findings are also applicable to other trace gases and aerosols. We explored relations between EFs and different satellite datasets thought to drive part of the variability in EFs (tree cover density, vegetation greenness, temperature, precipitation, and the length of the dry season). Although reasonable correlations were found for specific case studies, correlations based on the full suite of available measurements were less satisfying (r_{max} =0.62). This may be partly due to uncertainties in the driver datasets, differences in measurement techniques, assumptions on the ratio between flaming and smoldering combustion, and incomplete information on the location and timing of measurements. We derived new mean EFs, using the relative importance of each measurement location with regard to the amount of biomass burned. These weighted averages were within 18% of the arithmetic mean. We argue that from a global modeling perspective, future measurement campaigns could be more beneficial if measurements are made over the full fire season, or alternatively if relations between ambient conditions and EFs receive more attention.

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

To assess the atmospheric impact of biomass burning quantitatively, accurate emissions estimates of trace gases and aerosols are required. Crucial parameters include burned area, fuel consumption, and the emission factor (EF), usually defined as the amount of gas emitted per kg of dry fuel burned, expressed in units of g/kg dry matter (DM) (Andreae and Merlet, 2001, to which we will refer to as A&M2001).

Pioneering experiments to characterize fire emissions were conducted in South America (Crutzen et al., 1979), Africa (Delmas, 1982), and Australia (Ayers and Gillett, 1988). In the beginning of the 1990s, the experiments of these individual groups were followed by a number of large international campaigns of biomass burning experiments in various ecosystems throughout the world. These included the Southern Africa Fire-Atmosphere Research Initiative (SAFARI 92 and SAFARI 2000) in Southern Africa (Andreae et al., 1996a; Lindesay et al., 1996), Dynamique et Chimie Atmosphérique en Forêt Equatoriale-Fire of Savannas (DECAFE-FOS) in West Africa (Lacaux et al., 1995), Transport and Atmospheric Chemistry Near the Equator-Atlantic

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(Trace-A) over Brazil, Southern Africa, and the South Atlantic (Fishman et al., 1996), Fire Research Campaign Asia-North (FireSCAN) in Central Siberia (FIRESCAN Science Team, 1996), and Smoke, Clouds, and Radiation-Brazil (SCAR-B) in Brazil (Kaufman et al., 1998).

These coordinated studies and numerous independent smaller investigations have resulted in a large body of information on emission characteristics. Several summaries of experimental EF data were given based on available information at that time (e.g., Andreae, 1993; Delmas et al., 1995). The most extensive and frequently used summary is given by A&M2001, in which all the available data on fire emission characteristics for a large number of chemical species was synthesized into a consistent set of units. The measurements were stratified by biome type or fire use; tropical deforestation fires, savanna fires, extratropical forest fires, biofuel burning, charcoal making, charcoal burning, and agricultural residues. The database is updated annually (M. O. Andreae, personal communication, 2009).

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

In recent years, studies using satellite-retrieved trace gas concentrations have shown the potential to better constrain the estimates provided by bottom-up estimates such as GFED (Arellano et al., 2004; Edwards et al., 2004; Gloudemans et al., 2006). New burned area products (L3JRC (Tansey et al., 2007), MODIS (Roy et al., 2008; Giglio et al., 2010), GLOBCARBON (Plummer et al., 2006)) allow for a better characterization of the timing and locations of fire, although the quality of these burned products

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varies (Chang et al., 2009; Roy and Boschetti, 2009; Giglio et al., 2010). Combining bottom-up and top-down methods potentially allows for an assessment of the magnitude of emissions, as well as their spatio-temporal variability. This requires a thorough understanding of the relations between biomass combusted and emission of CO, CH₄, CO₂, and other trace gases or aerosols.

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

Here we evaluated existing information on EFs, based on an extensive database of field measurements (A&M2001; M. O. Andreae, personal communication, 2009), and explored several key factors determining part of the spatial and temporal variability in EFs. Satellite data on fraction tree cover, precipitation, temperature, Normalized Difference Vegetation index (NDVI), and length of the dry season were used to develop relations with the EFs for different vegetations types. We focused on CO, CH₄, and CO₂. However, since the Modified Combustion Efficiency (MCE), defined as the amount of carbon released as CO₂ divided by the amount of carbon released as CO₂ plus CO (Yokelson et al., 1996), has been used as an effective predictor for the emission of smoke gas composition from biomass fires (Ward et al., 1996; Sinha et al., 2003; Yokelson et al., 2003), our findings on CO and CO₂ EFs can be extended to better understand emissions of other trace gases and aerosols. We restricted our analysis

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to in-situ measurements due to the focus on spatio-temporal variability as a result of variability in vegetation and climatic conditions; laboratory measurements of EFs were not taken into account. We present new weighted EFs for specific vegetation types, and indicate how future EF experiments could be more beneficial from a global modeling perspective.

Fire processes

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

The combustion process

The combustion of the individual fuel elements proceeds through a sequence of stages (ignition, flaming, smoldering, and extinction), each with different chemical and physical processes that result in different emissions. Dry vegetation is made up of cellulose and hemicellulose (50-75%), which are the main constituents of the cell wall, lignin (15-35%), which binds the cellulose fibers and strengthens the cell wall, and different extractables (0-15%), which are organic species that are not part of the cellular structure of the biomass and can be dissolved or gassed out. Furthermore, vegetation consists of trace minerals (0-10%), and water.

The initial ignition is the phase before a self-sustaining fire can start, and it depends on both fuel (size, density, water content) and environmental (temperature, relative humidity, wind) factors whether the fuel is ignited or not. As long as the water content of the fuel is high, fuel temperature will remain too low for ignition due to evaporative cooling. Therefore, most of the water must be outgassed before ignition can occur. Besides water, organic volatiles are also released during this initial heating period.







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Once the fuel is sufficiently dry, combustion can proceed from the ignition phase to the flaming phase. It start with thermal degradation, in which water and volatile contents of the fuel are released, and is followed by the thermal cracking of the fuel molecules (pyrolytic step); high-molecular compounds are decomposed to char (less volatile solids with high carbon content), tar (molecules of intermediate molecular weight), and volatile compounds in the form of flammable white smoke. Several different compounds, with theoretically every possible molecule, are produced during this phase. This pyrolytic process starts when temperatures in the fuel bed are around 400 K. When temperatures exceed 450 K, the process becomes exothermic, and at 800 K (if oxygen is not limiting) glowing combustion begins. At this stage a complex mixture of tar and gas products are released, and when diluted with air, a flammable mixture may form. Flaming combustion occurs when this mixture ignites, and the complex mixture of mostly reduced substances emitted during pyrolysis are converted to simple molecules, particularly CO₂, H₂O, NO, N₂O, N₂, and SO₂ (A&M2001). Intermediate products like CO, CH₄, and H₂ may also be released during this stage, depending on the interaction between chemical kinetics and physical dynamics in the flame.

After most volatiles have been released and the rate of the pyrolysis slows down, less flammable compounds are produced; the flaming combustion ceases, and the smoldering phase begins. The major surface reaction in this phase is one of oxygen with carbon to form carbon monoxide. Smoldering combustion is a low-temperature process emitting large amounts of incompletely oxidized compounds, similar to the products of the initial solid phase decomposition during the flaming stage (Lobert et al., 1991; Yokelson et al., 1997). Hence this solid phase decomposition and the smoldering phase are responsible for the vast diversity of emission products. The amount of substances emitted from a given fire and their relative proportions are thus determined to a large extent by the ratio of flaming to smoldering combustion, often referred to as the combustion efficiency (CE).

Smoldering combustion can proceed for days even under relatively high moisture conditions if heat release and spread rate are balanced. When the flame is not present

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anymore during the smoldering process, the compounds are no longer oxidized but emitted as final products. This is the reason for the multiple low- or non-oxidized emissions, as well as low amounts of oxidized compounds such as CO₂ or NO. The slower rate of pyrolysis results in lower heat production and therefore in a lower decomposition rate, until the process terminates (extinction phase). Extinction can be influenced by convective cooling, a low oxygen supply, and changing fuel properties, for example when all fine fuels are combusted and fuel types with a low surface to volume ratio (for example stems) remain.

The combustion processes described above are somewhat simplified. For real-time open vegetation fires, different factors that influence the combustion process and which may change over time (e.g. meteorological conditions, differences in aboveground biomass density, topography) also need to be considered.

2.2 Factors influencing the EF

In general, the four most important factors controlling EFs of CO, CH₄, and CO₂ are vegetation characteristics, climate and weather, topography, and fire practices.

Vegetation characteristics 2.2.1

One of the most important factors influencing both the behavior and the emissions of a fire is the water content of the vegetation. The water content determines whether a plant or tree can ignite and what the burning efficiency will be. Vaporizing liquid water takes up a considerable amount of energy produced during a fire, and therefore the water in plants or trees (usually between 5% (dead savanna grasses during the dry season) and 200% (in fresh needles or leaves) of the vegetations' dry weight) has the capability to either stop a fire completely or to slow down the burning process (to a low smoldering stage). Thus, the main effect of fuel moisture on emissions is through a change in fire behavior; the enhancement of smoldering or reduction of flaming combustion, and hence, a reduction in overall burning efficiency (Lobert and

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Warnatz, 1993; Chen et al., 2010). Experiments of Lobert et al. (1991) showed that the ratio of CO/CO₂ increased by more than a factor two in specific burns, when the fuel moisture was increased from 5% to 16%.

Other important fuel characteristics related to vegetation are the size and density of the fuel consumed. Because fuel has to be heated to ignition temperature, small low-density fuel particles are more easily ignited than larger high-density particles. Once burning, the rate of heat production for smaller particles is higher than for larger particles, and therefore smaller particles are also capable of sustaining flaming combustion and supporting the burning of larger particles. Grass fuels in savannas have a large surface to volume ratio, are more easily oxidized, and therefore burn largely in the flaming phase, while stems, coarse litter, and organic soils that burn in forest fires are not as well oxidized and burn more in the smoldering phase. The propagation of fire in grass is much faster than on a trunk of wood, because less material has to be converted per unit volume.

2.2.2 Climate and weather

Climate plays an important role in the existence, settlement, and ecological support for vegetation, and thus determines the availability of fire fuel (Lobert and Warnatz, 1993). Fire frequency and the fire season are also partly determined by climatic factors. Fuels become more abundant and drier when the fire season (dry season) progresses. Korontzi et al. (2003) showed that the CO EF decreased from 100 g CO/kg DM in early dry season Africa savanna fires to 30 g CO/kg DM in peak dry season fires. A similar decrease in EF was observed for CH₄, while the CO₂ EF increased substantially.

Weather, on the other hand, has a more short-term impact on fire. Temperature, precipitation, and wind speed are factors that partly determine the occurrence of fires as well as their behavior, especially the CE. Temperature affects the fire probability and ignition due to its effect on fuel moisture. Precipitation is capable of inhibiting, completely stopping, or preventing a fire. Wind can have an effect on the spread rate of a fire, as fires usually propagate in two different directions; with the wind (heading fires)

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and into the wind (backing fires). Heading fires typically move faster, produce larger flames, but oxidize the fuel less completely than backing fires (Lobert and Warnatz, 1993). Therefore the production rate of emissions of incomplete combustion is higher for heading fires, and thus, the burning efficiency is lower. Lobert et al. (1991) showed an enhancement in the total relative emission ratio of CO over CO₂ from comparable savanna grass burns of 40%, if the burn changed from a backing to a heading fire.

2.2.3 Topography

Local topgraphy can change the burning behavior of a fire. Due to wind-channel effects, large fires in valleys or canyons show a significantly different spread rate compared to fires up a mountain or rim (Chandler et al., 1983). Smaller-scale fires are also heavily altered by sloping terrain. A burn behaves like a heading fire (as described in Sect. 2.2.2) if it proceeds upslope. A downslope fire behaves more like a backing fire, and thus burns more efficiently. This effect can be observed in different experiments where wind-driven fires are simulated by creating a slope for the burning platform (Lobert et al., 1991; Rothermel, 1967).

2.2.4 Fire practice

In the tropics and subtropics, fire has become for a large part a human-driven process. We expect that regional variations in fire practices influence EFs, especially in deforestation and agricultural fires. Slash and burn deforestation fires, for example, are very different from the burning of fuels that have been mechanically piled together into windrows and may burn more efficiently. This practice requires heavy machinery and is therefore limited to regions with more capital, for example the southern part of the Amazon where forests are cleared for soy production, amongst others (Morton et al., 2006). These different practices could impact EFs on regional scales. Added complexity is due to the practice of deforesting over drained peatlands in parts of Indonesia, thus including a fuel type with high EFs of reduced gases.

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3.1 Introduction

We used the EF database for different vegetation types that was compiled by A&M2001 and updated annually (M. O. Andreae, personal communication, 2009). The database consists of EFs measured during individual experiments, as well as during large international measurement campaigns. While the database also includes results from laboratory measurements, we excluded them and used only the results of ground and aircraft EF measurements. We did this because the focus of our work is on EF variability and the role of local (climatic) conditions, which are better represented by EF measurements in the field. Furthermore, A&M2001 also excluded laboratory measurements to calculate the biome-averaged EFs. Since we used the database of A&M2001, we also adopted their approach for converting emission data into EFs: most of the EFs in the database are measured using the carbon mass balance (CMB) method (Ward et al., 1979; Radke et al., 1990). The underlying premise of this method is that all carbon combusted in a fire is emitted into measurable portions in five forms: CO₂, CO, CH_4 , non-methane hydrocarbons (NMHC), and particulate carbon in smoke particles. The EF of a specie is then calculated from the ratio of the mass concentration of those species to the total carbon concentration emitted in the plume. To convert the EF to g/kg DM of fuel burned, the data need to be multiplied with the carbon content of the fuel. A&M2001 adopted a carbon content of 45% when this information was not given in literature cited.

When the emission data were given as molar emission ratios, the molecular weights of the trace and reference species were used to calculate the EF. Molar emission ratios can be obtained by dividing excess trace species concentrations measured in a fire plume by the excess concentration of a simultaneously measured reference gas (most often CO₂). If the EF of the reference species was not provided, the mean EF for the specific type of fire was used.

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With this database as a starting point, we compiled all EFs and searched the literature for accompanying ancillary data such as measurement location and timing. We then expanded the database to include location-specific parameters related to vegetation type and climate. We focused on the EFs of CO₂, CO, and CH₄ because these gases were measured during most campaigns, and the EF of CO₂ and CO can be used to calculate the modified combustion efficiency (MCE), which can be used to predict EFs of other species.

Available EF data

Figure 1 provides an overview of the locations where ground- and aircraft EF measurements were conducted for CO and CO₂. Fire emissions from the Global Fire Emissions Database version 3.1 (GFED3.1, Giglio et al., 2010; van der Werf et al., 2010) were used to identify the major biomass burning regions. Fire emissions were estimated based on satellite-derived burned area from the MODerate resolution Imaging Spectroradiometer (MODIS) sensor (Giglio et al., 2010) in combination with the Carnegie-Ames-Stanford Approach (CASA) biogeochemical model with a fire module (van der Werf et al., 2010).

Most locations with both CO and CO₂ EF measurements are in North America, the arc of deforestation in the Brazilian Amazon, Southern Africa (South Africa and Zambia), and Northern Australia (Fig. 1). While these areas are all major biomass burning regions, several other important regions lack measurements. These include Central Africa (e.g., Congo, Angola), Siberia, and Indonesia, although laboratory studies for Indonesian fuel samples exist (Christian et al., 2003). Each of these regions likely has relatively high rates of emissions of reduced gases; more woodland burning in Central Africa compared to Southern Africa where most savanna measurements were made, more groundfires in boreal Asia compared to boreal North America where most extratropical EFs were measured, and moister conditions and more peat burning in Indonesia compared to South America where most deforestation fire EFs were made.

To highlight the large variability in EFs, we plotted methane (CH₄) EFs against the molar MCE (based on CO and CO₂) in Fig. 2 for the three different vegetation biomes. 23570

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The biome-averaged EF values of A&M2001 and M. O. Andreae (personal communication, 2009) are also shown. In general, EFs in savannas and grasslands show high MCEs and a relatively low EF for CH₄, mainly because burning mostly takes place in the flaming phase. Tropical forest measurements on the other hand, show lower MCEs and higher values for the EF of CH₄, because these fires release more incompletely oxidized compounds. This is also the case for the extratropical forest measurements, although here the values are more variable. The correlation coefficient (r) between MCE and CH₄ for all in-situ measurements was -0.71 (y=-85.889x+85.278), and correlation coefficients for the different vegetation types were -0.80 (y=-61.447x+61.142), -0.81 (y=-104.551x+104.590), and -0.52 (y=-59.992x+60.967) for savanna and grasslands, tropical forest, and extratropical forest, respectively. Two extratropical forest measurements (Cofer et al., 1998 - MCE=0.78, EF CH₄=4.5; Hobbs et al., 1996 - MCE=0.81, EF CH₄=16.2) were excluded from this graph for clarity, but they were taken into account to calculate the correlation coefficient. Ward et al. (1992) and Hao and Ward (1993) have shown that the linear relationships between the MCE and EF of CH_4 are quite different for forest and savanna fires. This can also be seen in Fig. 2; the regression line between the CH₄ EFs and their MCE differs from the one derived for tropical forest.

The large spread apparent from Fig. 2 may be partly explained by the different control factors that we described in Sect. 2.2. One is related to the timing of the measurement, and thus to weather conditions during the fire (e.g., Korontzi et al., 2003). Fires in savannas and tropical forest areas usually burn during the late dry season, when fuels are abundant and dry enough to burn. Prescribed burning in tropical savannas on the other hand is often exercised in the early part of the dry season, and is commonly advocated when fire is used as a land management tool. Early season fires are less intense and result in a smaller amount of vegetation consumed and -probably more important- lead to less damage to the soil compared to late season fires. Pastoralists burn extensively in the early dry season to stimulate regrowth of palatable grasses for their cattle; fire is used for rapid nutrient release prior to the new growing season by

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

We explored the seasonal variation of the fire emissions for all EF data where a detailed description of the location and date of measurements was provided. To investigate whether the available measurements capture these seasonal variations we compared the carbon emissions for the EF measurement locations with the seasonal variation in carbon emissions according to GFED3.1. All locations where EF measurements were conducted for CO, CH₄, and CO₂, were used. Figure 3 shows the (cumulative) GFED3.1 fire carbon emissions for the month of maximum burning (MOMB – the month with the highest fire emissions in g C/month) for all the EF measurement locations in the savanna (dashed blue line) and tropical forest (dashed red line) biomes. For all the specific grid cells where EF measurements were taken, the month of highest fire emissions was used; the emissions for these particular months were added to define the MOMB. A strong seasonal cycle for both biomes is observed. The fire emissions for the month of EF measurements (MOM - the month when EF measurements were conducted) for savanna (blue line) and tropical forest (red line) measurement locations are also shown. Again, a seasonal cycle is observed. If EFs are measured instantenously instead of over a longer timeframe, EF measurements are usually taken during the peak of the fire season. Information on EFs during the shoulder of the season is then lacking, but may be an important contribution to total emissions in some areas. Ideally, EFs are thus measured during both peak and shoulder of the season. Comparing both lines (MOMB and MOM) indicates that most savanna and tropical forest measurements were conducted earlier in the fire season than would be expected when aiming to capture the full fire season. Overall the MOM cycle compares reasonably with the MOMB, and hence the EF measurements were conducted during the right month of the year. Extratropical forest measurements were excluded here, because the seasonal cycle in climatic conditions is not as pronounced and constant from year to year as in the tropics.

One of our main objectives was to explain the large variability in CO, CH_4 , and CO_2 EFs. For this, we compared all the EFs of the database with global monthly datasets of relevant parameters (as described in Sect. 2.2); fraction tree cover, precipitation, temperature, NDVI, and the length of the dry season. These parameters were chosen since global datasets are available for a longer period of time, although the spatial and temporal resolution is relative coarse (typically $0.5^{\circ} \times 0.5^{\circ}$ and monthly data) and may not fully capture key regional variability. Specific local and regional factors that may have a large influence on the EF variability, like e.g. wind, are excluded due to a lack of reliable data.

We used the fraction tree cover (FTC) product regridded to $0.5^{\circ} \times 0.5^{\circ}$ resolution for the year 2002 to represent the vegetation density and the ratio between grass and woody fuels in the EF measurement locations. The product was derived from the Vegetation Continuous Fields (VCF) collection which contains proportional estimates for vegetative cover types: woody vegetation, herbaceous vegetation, and bare ground (Hansen et al., 2003). The product was derived from seven bands of the MODIS sensor onboard NASA's Terra satellite. The continuous classification scheme of the VCF product better captures areas of heterogeneous land cover than traditional discrete classification schemes.

We used the 1°×1° daily (1DD) Global Precipitation Climatology Project (GPCP) precipitation product (Huffman et al., 2001) to estimate the precipitation dependence of EFs. This dataset is based on passive microwave measurements from the Special Sensor Microwave Imager (SSM/I), and infrared retrievals from the Geostationary Operational Environmental Satellite (GOES) and the Television InfraRed Observation Satellite (TIROS) Operational Vertical Sounder (TOVS). The monthly rainfall totals are corrected over some continental areas to match sparse ground-based observations, and at finer time scales the product relies exclusively on satellite-based precipitation estimates. We averaged the daily values to calculate a montly average (mm/month)

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for the years 1997–2008, the period of availability. For EF measurements conducted before the year 1997, we used the monthly 2.5°×2.5° GPCPv2.1 precipitation product (Adler et al., 2003), which is available from 1979 till present.

Temperature data were derived from a climatology and an anomaly source. The climatological data were downloaded from the Climate Research Unit (CRU) website (http://www.cru.uea.ac.uk/). We used the CRU CL 1.0 Mean Monthly Climatology product, with a resolution of $0.5^{\circ} \times 0.5^{\circ}$ (New et al., 1999). This dataset gives the mean monthly surface climate over global land areas, excluding Antarctica, and was interpolated from station data to 0.5° latitude-longitude for several variables. We then used the NASA GISS Surface Temperature Analysis (GISTEMP) as a source of temperature anomalies (Hansen et al., 1999). GISTEMP provides a measure of the global surface temperature anomaly with monthly resolution for the period since 1880, when a reasonable global distribution of meteorological stations was established. Input data for the analysis, collected by many national meteorological services around the world, is the unadjusted data of the Global Historical Climatology Network (Peterson and Vose, 1997). Documentation of the GISTEMP analysis is provided by Hansen et al. (1999), with several modifications described by Hansen et al. (2001). We used the 1961-1990 anomalies with a 1200 km smoothing radius, which were downloaded from the NASA website (http://data.giss.nasa.gov/gistemp/maps/). The CRU climatology and GISTEMP anomalies were combined to estimate the monthly temperatures for the years 1967-2009.

The Normalized Difference Vegetation Index (NDVI) represents the amount of live green vegetation and its productivity, and may be a useful indication of vegetation characteristics (fuel abundance and also moisture conditions). Monthly Global Inventory Modelling and Mapping Studies (GIMMS) NDVI data with a ~0.07°×0.07° resolution (Tucker et al., 2005) were downloaded from the International Satellite Land Surface Climatology Project website (http://islscp2.sesda.com/). Different satellite series of NOAA's Advanced Very High Resolution Radiometer (AVHRR) were used for this NDVI record. The dataset consists of bi-monthly NDVI data for the years 1981 to 2006.

which we averaged to monthly values. For EF measurements that were conducted before 1981 or after 2006, we used the monthly mean of the years 1981–2006.

The length of the dry season for the EF measurement locations was defined by counting the number of months in the 12 months before the measurement was conducted with precipitation rates below 100 mm/month (GPCP 1°×1° for the 1997–2009 period, and GPCPv2.1 2.5°×2.5° for 1979–1997). This parameter partly overlaps with the precipitation rates, but the added value lies in containing a memory of precipitation; it may be an indicator of the precipitation conditions before the month of the actual measurement. Small low-density fuels usually become faster in equilibrium with ambient moisture conditions than larger high-density fuels, which may require more than one month to dry out.

3.4 Correlations between driver data and EFs

In Table 1 the correlation coefficients between the driver data and the EFs of CO, CH_4 , and CO_2 are given. Here, we lumped all EF data for the three different biomes together. We performed simple linear regressions, with the EF as the dependant variable, and the different parameters that may control the EFs variability represent the independent variables. Besides the correlation coefficients (r), F-values were calculated to test if the regression between the EF and the different driver data was significant (if the F-value exceeds the critical value of F_{crit} , it indicates a significant fit).

Besides the linear regressions, we also performed a multivariate regression to construct a regression equation that combined the different parameters that accounted for most of the EF variability, in order to see if different variables combined perform better than the variables seperately.

For CO EF we found the highest correlation with FTC (r=0.49) and NDVI (r=0.41). The corresponding F-values (66.2 and 7.0) exceeded the critical F value (F_{crit}=6.7) for a significance level of 0.01. When combining the different parameters in one regression equation, the correlation coefficient improved to 0.57. For the CH₄ EF, FTC (r=0.58) and precipitation (r=0.53) were the most dominant parameters, and both correlations

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were significant at a significance level of 0.01. Using the additional information of each parameter increased the correlation (r=0.62). For CO₂, FTC and precipitation yielded the highest descriptive power (r=-0.26 and r=-0.37), just like for CH₄. Despite the relatively low correlation coefficients, both fits were significant with F-values of 10.1 and 27.1. The multivariate regression equation gave a slightly higher correlation (r=0.43). Repeating the calculations but focusing on biome-specific EFs yielded lower correlations than with all measurements lumped together. Especially correlations between the EFs and the driver data for the extratropical forest were very poor. Possible explanations for these poor correlations are discussed in Sect. 4. Higher correlations between EFs and the driving variables were found when focusing on specific locations. Figure 4a–c shows correlations for, respectively Brazilian deforestation fires and savanna fires in Australia (FTC vs. MCE), Brazilian deforestation fires (FTC vs. CH₄EF), and boreal fires in Alaska (precipitation vs. CH₄EF).

3.5 Weighted EF averages

Most large-scale biomass burning emission estimates are based on some combination of biomass or carbon combusted and EFs. These EFs are usually based on the arithmetic mean of a large number of measurements, most often based on the work of A&M2001 with annual updates (M. O. Andreae, personal communication, 2009). It is not known, however, whether the measurements are representative of the whole biome. Regionally, there is substantial variation in the density of measurements. For example, nearly all tropical forest measurements are made in the Brazilian Amazon (Fig. 1), while information from other deforestation hot spots such as Bolivia and Indonesia is lacking. The same holds for the boreal region; according to the estimates of van der Werf et al. (2010), total carbon emissions from boreal Asia were almost 2.5 times as high as those from boreal North America in the last decade. Nevertheless nearly all the extratropical forest EF measurements were made in North America, and only one was conducted in boreal Asia (Fig. 1).

While there are regional discrepancies in measurement locations, the measurements do capture most of the climate window in which most fires occur (Fig. 5). To construct

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new weighted average EFs, we weighted each measurement with its role in the fire-climate window. The size of the climatic window bins we used were 1 °C for mean annual temperature (MAT), 100 mm/year for mean annual precipitation (MAP), and 2% for fraction tree cover (FTC). Table 2 gives an overview of these new calculated mean values per biome. The weighted values are within 18% of the arithmetic mean (Table 2). Some differences, however, can be noticed: EFs of CO were 8% below and 13% above the mean of A&M2001 for tropical forest and extratropical forest measurements, respectively. EFs of CH₄ were lower for each biome (16% on average). CO₂ EFs were somewhat lower for savannas (1.5%) and more variable for the tropical and extratropical biome.

4 Discussion

We evaluated available literature describing EF measurements conducted in different biomes throughout the world, and explored the relations between the EFs and global low-resolution datasets of parameters that influence EF variability. We chose to compare EFs with five important control parameters for which global datasets were available and extended back to at least the early 1990s. These could account for up to about 32% (r=0.57), 38% (r=0.62), and 18% (r=0.43) of the variability for, respectively CO, CH₄, and CO₂. Several factors may account for the remaining variability and are discussed in Sects. 4.1–4.4. We discuss the new weighted biome-averaged EFs in Sect. 4.5, followed by recommendations for new EF campains (Sect. 4.6) and our future steps (Sect. 4.7).

4.1 Driver data uncertainty

Monthly averages of coarse-resolution (~0.07°, 0.5° and 1.0°) data were used to represent fire emissions, fraction tree cover, precipitation, temperature, NDVI, and the length of the dry season for the different EF measurement locations. The use of spatially and

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temporal higher resolution data is preferred over lower resolution data, but detailed information on location and date of the measurements was often lacking. Even if detailed information was given, a large number of EF measurements were conducted in the 1980s and early 1990s, for which period global datasets are often lacking at sufficient high resolution.

4.2 Additional emission factor drivers

Although specific case studies have proven that wind and topography plays an important role in fire characteristics and thus in the partitioning of trace gases emitted (e.g., Lobert et al., 1991), we have not taken these factors into account because reliable information is simply not available from global datasets. Few papers describing the measurements include detailed information on climatic and environmental conditions. Fuel composition and its moisture content are other crucial factors for EF partitioning which were not taken into account here, and which may account for part of the variability not captured by the 5 parameters we included and for which consistent information was available for all measurement locations.

4.3 Different measurement approaches and techniques

Various analytical techniques have been used in recent field experiments, like non-dispersive infrared analysis (NDIR), Fourier transform infrared spectrospocy (FTIR), and gas chromatography. Detailed descriptions of these different techniques can be found in the literature (Ward and Radke, 1993; Yokelson et al., 1999; Christian et al., 2004). For real-time concentration measurements, the analytical instruments must be close to the fire. A distinction can be made between ground-based (tower, mast) and airborne (airplane, helicopter) measurements. Airborne measurements sample an integrated mixture of the emissions from both combustion types (smoldering and flaming). For ground-based measurements, which have a smaller footprint, the separation between smoldering and flaming combustion is more clear, but even here both processes occur simultaneously in a given patch at most times. Ground-based sampling probably

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oversamples the emissions which tend to be emitted during less vigorous phases of a fire and therefore to remain closer to the ground, while airborne sampling may be biased towards emissions from the flaming phase that rise to higher altitudes (Andreae et al., 1996b).

Airborne measurements of chapparral vegetation in California (Laursen et al., 1992) were for example compared to ground-based measurements of the same vegetation type (Ward and Hardy, 1989), with overall lower EFs for CO (18%) and CH $_4$ (60%), and higher CO $_2$ (5%) due to the bias towards the flaming phase. The height of the plume where samples were taken may also play an important role; the higher the sampleheights, the stronger the bias towards emissions from the flaming phase.

Not only the measurement location impacts the measurements, also the different techniques used to sample the smoke may yield variability that we cannot account for. In general, two systems are used: the Fire Atmospheric Sampling System (FASS) (Ward et al., 1992) consists of a mast set up on experimental plots before a fire. The mast is equipped with temperature and wind probes, and gas and particulate matter samplers are connected to real-time gas (CO₂, CO, NO) sensors or canisters placed in a computer-driven data logger module that is inserted in the soil at the bottom of the mast. FASS, which has been used in South-America and Africa, provides a single integrated measurement of the whole fire process in each studied plot, and can be used for any type of fire, even very intense ones. Another system, used in the FOS/DECAFE and SAFARI experiments, yields continuous measurements because it follows the fire front. It is based on handheld sampling devices supplied by tubing held on long poles and connected to real-time gas analyzers. Canisters or Teflon bags for laboratory analyses can be filled using the same system.

The use of different techniques may cause variation in the EFs measured. For example, SAFARI campaign measurements were conducted in South Africa and Zambia, and different research groups were involved to estimate EFs. Airborne Fourier transform infrared spectroscopy (AFTIR) was used by Yokelson et al. (2003) to measure EFs, while Sinha et al. (2003) used gas chromatography. Both measuring techniques

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gave different EFs of CO, CH_4 and CO_2 , even though the same plumes were sampled. Another example comes from extratropical forest biome; the use of different analystical techniques led to a difference of 22.7% for CO, 8.3% for CH_4 , and 2.2% for CO_2 EFs for the same fires in North America (Hegg et al., 1990; Laursen et al., 1992).

Clearly, this measurement-related variability is large but cannot be accounted for, and these shortcomings should be considered when comparing EF measurements.

4.4 Flaming/smoldering assumptions

The ratio between flaming and smoldering combustion of a fire is crucial for estimating the overall EF for different trace gases. In savanna fires, for example, flaming combustion dominates, and the EF for reduced species is relatively low compared to forest fires where the smoldering fase is often more important. The proportion of flaming and smoldering combustion can vary considerably also within fires in the same biome as a function of internal parameters (for example moisture content). It may seem desirable to provide separate EFs for flaming and smoldering combustion, but this is not always possible given the data available. In the field, EFs are generally determined by averaging several instantaneous measurements from the fire. Most emissions are assumed to be a mixture of flaming and smoldering combustion, and it is essential that averaging of both phases is done correctly when the EF for an entire fire is sought. Generally the individual measurements are weighted according to the amount of fuel combusted in the time interval represented by the measurement (Ward and Hardy, 1991). This approach requires information that is only available in experimental fires in the laboratory or to a limited extent in the field.

Estimates of the relative importance of the flaming and smoldering phases vary in literature; for grass and shrub fires flaming combustion dominates and likely accounts for 80% to 90% of fuel consumption (Shea et al., 1996; Ward et al., 1996). For tropical forest and boreal fires smoldering combustion is more important. Bertschi et al. (2003), for example, assumed that the smoldering and flaming phases combusted equal amounts of biomass in boreal areas, and residual smoldering measurements were combined

with airborne measurements of Goode et al. (2000) to calculate an overall EF. For African miombo fires a flaming-smoldering ratio of 90–10 was taken, and airborne FTIR measurements from a study of Yokelson et al. (2003) were used to represent the flaming part. A change in these flaming-smoldering ratio's will impact the overall EF substantially, so the assumptions made by different authors are therefore important to consider.

A&M2001 made the assumption that when smoldering and flaming emissions were given separately in ground-based studies, the emissions were combined to represent the complete fire. For this purpose A&M2001 either used data on the fractions of fuel combusted in the smoldering and flaming stages provided in a given study, or, when this information was not available, typical values from other studies on the same type of fire.

4.5 Weighted means

The biome-averaged EF values of A&M2001 and M. O. Andreae (personal communication, 2009) are widely used in the modeling community. Although differences (Sects. 4.3 and 4.4) exist between all the measurements, all data were averaged to calculate a mean EF per vegetation biome. These mean values may not always be representative for the whole biome (e.g. nearly all extratropical forest measurements were made in North America, and only one measurement was made in Russia). By placing the measurements in their climatic window (based on mean annual precipitation, mean annual temperature, and fraction tree cover) we were able to weigh the different measurements, using the GFED3.1 carbon emissions estimates in the corresponding carbon climatic window. The weighted EFs are within 6.7%, 7.9%, and 13.2% of the arithmetic mean of A&M2001 for CO, and 17.4%, 15.2%, and 6.7% for CH₄ for the savanna, tropical forest, and extratropical forest biome, respectively. The weighted EFs of CO₂ are within 3% of the arithmetric mean for all three biomes. According to the linear regression results for the different EF drivers, the climatic window with the most predictive power for CO, CH₄ and CO₂ EFs together is based on fraction tree

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cover and mean annual precipitation (Table 2, FTC-MAP). Based on the weighting by FTC and MAP, the EFs are systematically lower than the arithmetic mean of A&M2001, with a 8.7%, 3.7%, and 2.1% decrease for CH_4 , and 1.2%, 1.5%, and 0.4% for CO_2 , for the savanna, tropical forest, and extratropical forest biome, respectively. For CO the weighted EFs were lower than the arithmetic mean of A&M2001 for savanna and tropical forest (1.7% and 7.9%), but higher for extratropical forest (3.8%).

We adopted the different vegetation types that were defined by A&M2001, and based on these biomes (savanna and grasslands, tropical forest, extratropical forest), we calculated new weighted EF averages. Specifically, several measurements were conducted in vegetation types (for example chaparral in California and pinetree forest in Mexico) that cannot be clearly classified as savanna and grassland, tropical forest, or extratropical forest. While the savanna and tropical forest biome EF measurements were clustered in Fig. 5, the extratropical forest measurements show more variation (especially Fig. 5b). For a more specific EF average, it could be helpful to expand the amount of vegetation types, for example by adding a "temperate forest" biome.

However, instead of a discrete classification based on a limited number of biome types, the large spread in EFs may be better represented by stratifying EFs by vegetation density (FTC, NDVI) and climatic conditions (precipitation, temperature, length of dry season).

4.6 Recommendations for future EF campaigns

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

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

Seasonality: to better understand the temporal variation of EFs in specific vegetation biomes, there is a need of measurements made over the full fire season, following Korontzi et al. (2003).

Fuel and ambient conditions: measuring and describing fuel composition, its moisture content, and ambient conditions such as windspeed and temperature may allow for a better understanding of the factors driving EFs, especially when multiple locations are visited with the same measurement protocol. This requires a more multi-disciplinary approach and calls for combining campains aiming to quantify biomass loads, combustion completeness, and EFs.

4.7 Future steps

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

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The partitioning of combusted biomass into trace gases and aerosols shows large variation in time and space. We assessed what fraction of this variability can be explained by coarse resolution, globally available datasets including fraction tree cover, precipitation, and temperature. When combined, these datasets could account for up to about 38% (r=0.62) of the variability in emission factors. Uncertainties in driver data, differences in measuring techniques, assumptions on weighting ratios of flaming and smoldering contributions, and insufficient information on the measurements may account for part of the remaining variability. The same holds for neglected driver data such as topography and windspeed.

We have calculated new average emission factors for three biomes, by weighting the EFs with the amount of biomass combusted. Using the climatic window with the highest predictive power, weighted EFs are lower than the arithmetric mean of A&M2001, with a 8.7%, 3.7%, and 2.1% decrease for CH_4 , and 1.2%, 1.5%, and 0.4% for CO_2 , for the savanna, tropical forest, and extratropical forest biome, respectively. For CO the weighted EFs were lower than the arithmetic mean of A&M2001 for savanna and tropical forest (1.7% and 7.9%), and higher for extratropical forest (3.8%).

Although 70 different papers are found in literature describing emission factor measurements that have been made in various biome types for CO, CH₄, and CO₂, most lack a detailed description of the measurement site. In the future, a 1) stronger focus on factors governing the variability in emission, 2) exploring relatively uncharted areas including Central Africa, boreal Asia, and Indonesia, all regions with likely higher-than-averge EFs, and 3) performing measurements over the full fire season may lead to a better understanding of the variability in emission factors. The development of a more uniform sampling protocol, for the sampling and measurements of EFs in different vegetation types, may be helpful to better compare different measurements.

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

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

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Table 1. Correlation coefficients (r) and F-values (F) for CO, CH₄, and CO₂ EF measurements and different driver data. The correlation coefficient for the multivariate regression equation is also shown (r combined). n corresponds to the number of samples used, and F-values shown in Italic indicate relations that did not exceed the critical F-value for a significance level of 0.01.

Driver data	CO (n=216)		CH ₄ (n=205)		CO ₂ (n=169)	
	r	F	r	F	r	F
Fraction tree cover	0.49	66.2	0.58	104.3	-0.26	10.1
Monthly precipitation	0.40	1.9	0.53	13.8	-0.37	27.1
Monthly temperature	-0.13	0.1	0.03	0.1	-0.13	2.7
Monthly NDVI	0.41	7.0	0.39	0.5	-0.22	0.2
Length dry season <100 mm	0.19	20.1	-0.04	0.6	0.02	5.9
r combined	0.57		0.62		0.43	

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Table 2. EFs of CO, CH_4 , and CO_2 for savanna (S), tropical forest (T), and extratropical forest (E), weighted by carbon emissions and stratified by mean annual precipitation (MAP), mean annual temperature (MAT), and fraction tree cover (FTC) bins. Biome-averaged arithmetic means of A&M2001 and M. O. Andreae (personal communication, 2009) are also shown, with standard deviations in parenthesis. The results for the climatic window with the highest predictive power are shown in Italic.

	CO			CH ₄			CO ₂		
	${\mathcal S}$	Τ	Ε	$\mathcal S$	T	Ε	$\mathcal S$	T	E
MAP-MAT	56	94	107	1.9	5.6	4.0	1624	1636	1588
FTC-MAT	61	97	120	2.1	5.8	4.7	1622	1615	1529
FTC-MAP	59	93	112	2.1	5.7	4.7	1627	1601	1565
A&M(2001)	60 (19)	101 (16)	106 (36)	2.3 (0.8)	6.6 (1.8)	4.8 (1.8)	1646 (99)	1626 (39)	1572 (106)

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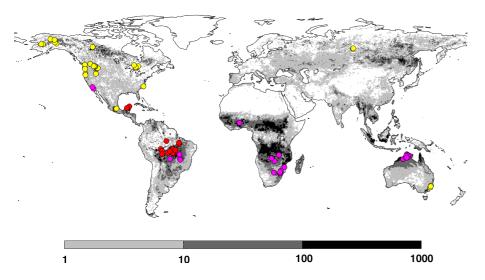


Fig. 1. Locations where CO and CO₂ EFs were measured. Locations were stratified by biome following A&M2001; savanna and grassland (purple), tropical forest (red), and extra-tropical forest (yellow). Background map shows annual GFED3.1 fire emissions in g C/m²/year, averaged over 1997-2009, and plotted on a log scale.

Discussion Paper

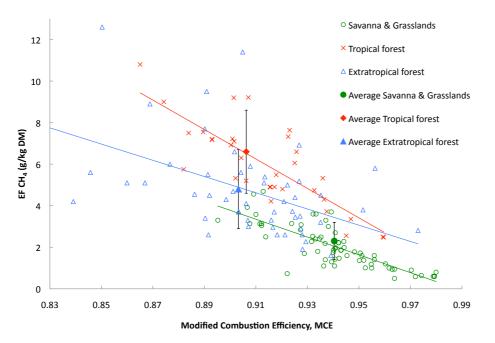


Fig. 2. Methane (CH_4) EFs and the molar-based modified combustion efficiency (MCE) for all available measurements including the biome-averaged values presented in A&M2001 and M. O. Andreae (personal communication, 2009) and regression lines. The errorbar indicates the reported standard deviation.

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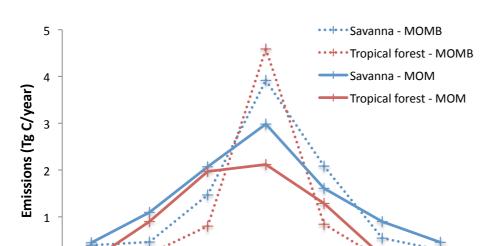


Fig. 3. GFED3.1 fire emissions in Tg C/year (mean for 1997–2009) for all savanna (solid blue line) and tropical forest (solid red line) EF measurement locations. MOM corresponds to the month when EF measurements were conducted. The dashed line shows the emissions for the month of maximum burning (MOMB), and the months before and after this month.

MOMB

Month

1

2

3

-1

0

-3

-2

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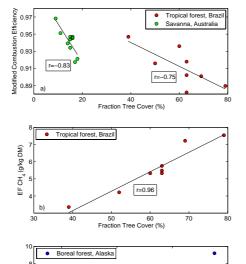


Fig. 4. Relations between driver data and EFs or MCE for selected regions. **(a)** fraction tree cover and modified combustion efficiency (MCE) for savanna measurements in Australia (Hurst et al., 1994; Shirai et al., 2003) and tropical deforestation measurements in Brazil (Yokelson et al., 2007), **(b)** fraction tree cover and CH_4 EF for tropical deforestation measurements in Brazil (Yokelson et al., 2007), and **(c)** precipitation and CH_4 EF for extra-tropical forest measurements in Alaska (Laursen et al., 1992; Goode et al., 2000; Wofsy et al., 1992; Nance et al., 1993).

GPCP precipitation (mm/year)

r=0.88

650

CH₄ (g/kg DM)

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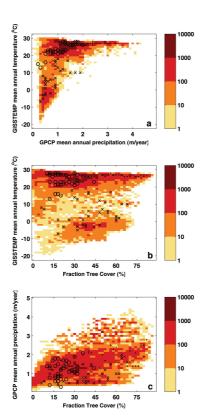


Fig. 5. GFED3.1 fire emissions in 1E10 g C/year (mean for 1997–2009) in a temperature – precipitation (a), temperature - fraction tree cover (b), and precipitation - fraction tree cover (c) window overlain by EF measurements in savanna and grasslands (circles), tropical forest (dots), and extratropical forest (crosses). Temperature and precipiation were averaged over 1990-2008. 23599

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