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Biomass burning aerosol emissions from vegetation fires: particle number and mass emission factors and size distributions

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Abstract

Aerosol emissions from vegetation fires have a large impact on air quality and climate. In this study, we use published experimental data and different fitting procedures to derive dynamic particle number and mass emission factors (EF_{PN}, EF_{PM}) related to the fuel type, burning conditions and the mass of dry fuel burned, as well as characteristic CO-referenced emission ratios (PN/CO, PM/CO). Moreover, we explore and characterize the variability of the particle size distribution of fresh smoke, which is typically dominated by a lognormal accumulation mode with count median diameter around 120 nm (depending on age, fuel and combustion efficiency), and its effect on the relationship between particle number and mass emission factors.

For the particle number emission factor of vegetation fires, we found no dependence on fuel type and obtained the following parameterization as a function of modified combustion efficiency (MCE): $EF_{PN}=34{\cdot}10^{15}\times(1\text{-MCE})\,kg^{-1}\pm10^{15}\,kg^{-1}$ with regard to dry fuel mass (d.m.). For the fine particle mass emission factors (EF_{PM}) we obtained $(86–85\times MCE)\,g\,kg^{-1}\pm3\,g\,kg^{-1}$ as an average for all investigated fires; $(93–90\times MCE)\,g\,kg^{-1}\pm4\,g\,kg^{-1}$ for forest; $(67–65\times MCE)\,g\,kg^{-1}\pm2\,g\,kg^{-1}$ for savanna; $(63–62\times MCE)\,g\,kg^{-1}\pm1\,g\,kg^{-1}$ for grass.

For the PN/CO emission ratio we obtained an average of $(34\pm16)\,cm^{-3}\,ppb^{-1}$ exhibiting no systematic dependence on fuel type or combustion efficiency. The average PM/CO emission ratios were $(0.09\pm0.04)\,g\,g^{-1}$ for all investigated fires, $(0.13\pm0.05)\,g\,g^{-1}$ for forest; $(0.08\pm0.03)\,g\,g^{-1}$ for savanna; and $(0.07\pm0.03)\,g\,g^{-1}$ for grass.

The results are consistent with each other, given that particles from forest fires are on average larger than those from savanna and grass fires. This assumption and the above parameterizations represent the current state of knowledge, but they are based on a rather limited amount of experimental data which should be complemented by further measurements. Nevertheless, the presented parameterizations appear sufficiently robust for exploring the influence of vegetation fires on aerosol particle number

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and mass concentrations in regional and global model studies.

1 Introduction

Aerosol particle emissions from vegetation fires have large impacts on both climate and air quality (Yokelson et al., 2007; Andreae and Crutzen, 1997; Andreae et al., 2004). During burning periods, the visibility in affected areas can be heavily reduced, and the health effects on the local populations can be substantial. Biomass burning particles are efficient cloud condensation nuclei (CCN) and can influence the formation of clouds and precipitation (Luderer et al., 2006; Trentmann et al., 2006; Kivekäs, 2008; Reid et al., 2005; Reutter et al., 2009; Roberts et al., 2002; Rissler et al., 2004, Feingold et al., 2001; Asa-Awuku et al., 2008; Rose et al., 2008; Rosenfeld et al., 2008).

There is a growing interest in the indirect aerosol effect in climate models, but to fully represent the effect of aerosol emissions on the cloud properties, improved particle number emission factors are needed (Andreae and Rosenfeld, 2008; Fuzzi et al., 2006; Lohmann et al., 2007). Currently, emission factors are mainly related to fuel types, but as the understanding of the fire process increases, the emission factors are not just treated as pure averages over the fire but can be related to fire properties as well (Hu et al., 2008; van der Werf et al., 2006; Schultz et al., 2008; Thonicke and Cramer, 2006; Hodzic et al., 2007). In this way, changes in the fire process due to, e.g., meteorological effects can also be taken into account in the models.

Particle emissions from biomass burning are dominated by an accumulation mode, with a count median diameter of 100–150 nm, together with two smaller modes; a coarse mode, and occasionally also a nucleation mode (Reid et al., 2005). The composition of the particles depends both on the fuel and on the burning process. The coarse mode particles consist of dust, carbon aggregates, ash and unburned parts of the fuel (Hungershoefer et al., 2008; Formenti et al., 2003; Gaudichet et al., 1995), while the accumulation mode consists mostly of organic matter, with soot carbon and inorganic species making up ~10% each (Reid et al., 2005). Of the organic matter 40–

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80% is water soluble and 20–40% consists of acids (Reid et al., 2005), while alcohols and sugars, e.g., levoglucosan, make up less than 5% of the organic matter (Oros et al., 2006).

In the size range between the coarse and the accumulation modes, at a particle diameter around 1 μm, the emissions of both particle number and particle mass are minor (Radke et al., 1991; Falkovich et al., 2005; Hardy et al., 1996; Hays et al., 2005). This study focuses on the accumulation mode with a count median diameter around 120 nm and a mass median diameter around 240 nm, which includes most of the particles, both by number and mass (Reid et al., 2005).

The particle size distribution of biomass burning emissions is extremely dynamic in the initial plume. Close to the fire, i.e., less than a few minutes away, a nucleation mode is often present. It is mainly detected in laboratory studies (Hays et al., 2005; Wardoyo et al., 2006; Keshtkar and Ashbaugh, 2007), but also in the field (Formenti et al., 2003; Sinha et al., 2003). These particles can be numerous, but have almost no mass and little influence on aerosol optical properties and CCN activity. Normally at a timescale of minutes up to half an hour, the nucleation mode particles transfer into the accumulation mode.

Compared to the accumulation mode, the coarse particles are few, but can make up a significant fraction of the particle mass. Most of the data show a limited amount of coarse particles in the biomass plume (Reid et al., 2005; Schafer et al., 2008), e.g., a ratio of PM_{10} to $PM_{2.5}$ of 1.3±0.2 for vegetation fire plumes compared to 2.4±0.5 for background conditions in Montana (Ward et al., 2006). Radke et al. (1991) found that, on average, the coarse mode accounts for about 20% of the mass of smoke aerosol emitted. Particles in smoke plumes can reach quite large sizes: Instrumental observations show continuous log-normal size distributions reaching the millimeter size range (Radke et al., 1990; D. Rosenfeld, unpublished data, 2002), and visual observations of the fallout below fire plumes frequently show centimeter- to meter-sized objects (ash, char, burning branches, etc.).

The aim of this study is to parameterize the emission of biomass burning particles

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from vegetation fires. We have compiled all available literature data, present particle number and mass emission factors and ratios, and relate these to combustion efficiency and fuel type. We have analyzed three fuel types; forest, savanna and grass. Particle number and particle mass emissions are described separately and related through particle size distributions. This gives a consistency check on the results obtained, and also provides a starting point for the continuation of emission studies, both theoretical and experimental.

2 Definitions and methods

The emission factor (EF) is defined as the amount of aerosol particles that are emitted per kg of dry fuel mass burned; and is measured either as particle mass (EF_{PM}) or as particle number (EF_{PN}). To estimate the amount of fuel in an open vegetation fire, where weighing of the fuel is not possible for practical reasons, the common approach is to measure the various carbon species in the smoke. The assumption is made that all carbon in the burned part of the fuel is found in the smoke, and generally that the carbon content of the fuel is 45% of the mass (Andreae and Merlet, 2001). The other approach used is to scale the particle emissions to carbon monoxide, and to present CO-referenced emission ratios (PN/CO, PM/CO). PN/CO usually refers to the particle number concentration at 100 kPa and 298 K divided by the CO concentration or volume mixing ratio, respectively (common unit: $cm^{-3}\,ppb^{-1}$), while PM/CO is the ratio of particle and CO mass concentrations (common unit: $g\,g^{-1}$).

The combustion efficiency (CE) of a fire is generally defined as the amount of carbon released in the form of carbon dioxide divided by the total amount of carbon released. In many cases only CO and CO_2 are measured, and the modified combustion efficiency (MCE) is used to characterize burning conditions. The MCE is defined as the amount of carbon released as CO_2 divided by the amount of carbon released as CO_2 plus CO (Yokelson et al., 1996, Eq. 1). The difference between MCE and CE is normally less than a few percent (e.g. Guyon et al., 2005), and for this study MCE will be used

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exclusively to increase comparability.

$$MCE = (\Delta C_{CO_2})/(\Delta C_{CO_2} + \Delta C_{CO}) \quad (1)$$

The particle mass is measured as the collective mass of all particles below a given size limit, shown here as a subscript to PM (e.g., $PM_{3.5}$ for particles smaller than 3.5 μm), in analogy with the conventionally defined PM_{10} and $PM_{2.5}$.

Linear fitting methods have been used to find relationships between particle emissions and MCE, and between other parameters in the smoke. Following the recommendations of Cantrell (2008), standard linear least squares fitting was used for the parameterization of EF as a function of MCE, whereas the bivariate fitting method described in Cantrell (2008) was used to relate the geometric mean diameter to the geometric standard deviation of the lognormal size distribution of smoke particles. F-statistics has been used to verify relationships found between parameters. From the F-value and the df value, the probability of erroneously finding a relationship between two factors of interest, P_{err}, is calculated using the LINEST and FDIST functions of Microsoft EXCEL.

3 Particle size distribution

Biomass burning emissions are mainly in the accumulation mode, and can be described by a lognormal size distribution (Hinds, 1998; Seinfeld and Pandis, 2006) with a count median diameter, D_g (similar to the geometric mean diameter). The fresh smoke arithmetic mean ± standard deviation is D_g=(117±13) nm and the average geometric standard deviation of the particle size distribution is σ_g=1.7±0.1 (the number of data point in the average, n=20, Reid et al., 1998; Guyon et al., 2005; Reid and Hobbs, 1998). For aged smoke D_g=235±40 nm, σ_g=1.4±0.1 (n=14; Anderson et al., 1996; Fiebig et al., 2003; Formenti et al., 2002; Petzold et al., 2007; Reid et al., 1998, Table S1 in the online Supplement http://www.atmos-chem-phys-discuss.net/9/17183/

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2009/acpd-9-17183-2009-supplement.pdf). Both D_g and σ_g vary with MCE and are interrelated.

3.1 Relationship between D_g and MCE

Particles emitted during flaming combustion are commonly larger than those emitted during smoldering combustion (Reid and Hobbs, 1998; Hobbs et al., 1996; Rissler et al., 2006; Hays et al., 2002; Wardoyo et al., 2006). Under very strongly smoldering conditions, particles seem to become larger again, but this applies mainly to peat fires at very low combustion efficiencies below 0.7 (Iinuma et al., 2007). The particle size relation to MCE is a linear fit on fresh Brazilian forest smoke (MCE=0.85–0.98, age<4 min, number of data points n=11, correlation coefficient R^2=0.83, Reid and Hobbs, 1998):

$$D_g/[\text{nm}] = 240 \times \text{MCE} - 100 \quad (2)$$

3.2 Relationship between D_g and σ_g

Figure 1 shows a compilation of published data of D_g and σ_g for fresh and aged biomass burning smoke from vegetation fires. Fresh means smoke plumes younger than ~1 h; aged smoke data are mostly from plumes older than one day (Table S1 in Supplement http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf).

The smoke data have been fitted linearly with a bivariate method (Cantrell, 2008), as errors exist in both x and y; and they co-vary without a causative relationship between x and y for fresh, aged, and all data, respectively (Eqs. 3–5). The standard fitting method gives different relations depending on the direction of the fit, i.e., if D_g or σ_g is at the x-axis. One of these fits gives approximately the same result as the bivariate fit, while the relation that differs from the bivariate relation gives less variation in D_g with similar variation in σ_g, see Eqs. (S3–S5) in Supplement (http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf). The correlation between D_g

and σ_g increases when including data for aged smoke (R^2=0.30, n=20, Eq. (3) for fresh smoke, R^2=0.52, n=14, Eq. (4) for aged smoke and R^2=0.80, n=34, Eq. (5) for the overall data set). The fresh data is the focus of this study and thus Eq. (3) will be used in further analysis, if not stated otherwise.

$$\text{Fresh}: D_g/[\text{nm}] = (584 \pm 5) - (269 \pm 1) \times \sigma_g \quad (3)$$

$$\text{Aged}: D_g/[\text{nm}] = (784 \pm 11) - (382 \pm 1) \times \sigma_g \quad (4)$$

$$\text{Alldata}: D_g/[\text{nm}] = (797 \pm 6) - (392 \pm 1) \times \sigma_g \quad (5)$$

4 Particle number emissions

4.1 Literature data and average values

Table 1 gives an overview of studies reporting aerosol particle number emission factors (EF_{PN}) and CO emission ratios (PN/CO) from field measurements of biomass burning smoke plumes released by vegetation fires. Very freshly emitted smoke usually contains large amounts of nucleation mode particles with diameters <30 nm, but they are rapidly lost by coagulation on time scales of minutes and have little influence on the large scale properties and effects of atmospheric aerosols and clouds (Andreae and Rosenfeld, 2008; Hobbs et al., 2003). Thus, the very high EF_{PN} and PN/CO values from studies investigating very fresh smoke within the first few minutes after emission (Table 1: Sinha et al., 2003) are not included in our further analysis. The three last studies listed in Table 1 refer to aged smoke, and also have rather high lower particle size cutoff (~120 nm), and are thus not used in the further analysis. The available data is limited to flaming conditions, i.e., MCE larger than 0.9 (the study-averaged MCEs were between 0.93 and 0.97).

To make the numbers comparable, in spite of the different lower size limits, the Le Canut et al. data is assumed to have the same size distribution and lower particle size

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cut off as the Formenti et al. data measured in the same area using similar instrumentation. For conversions between the emission ratio [cm^{-3} ppb^{-1}] and the emission factor [kg^{-1} d.m. for PN and g kg^{-1} d.m. for CO] we assumed 298 K and 100 kPa.

The averaged emission factors for accumulation mode particles, not taking the MCE relationship into account and using size limited corrected overall data, was $(1.7\pm1.2)\cdot10^{15}$ kg^{-1} d.m., with forest fire emission factors of $(1.9\pm1.3)\cdot10^{15}$ kg^{-1} d.m. The particle number to CO emission ratios for the overall data set was 34 ± 16 cm^{-3} ppb^{-1}, with 30 ± 14 cm^{-3} ppb^{-1} for forest fires, that can be compared to savanna and grass type fuels in Table 1. The data suggests a larger emission factor and a smaller emission ratio for the forest emissions, but the differences between the fuels are within the standard deviation of each measurement, and we cannot show that the difference is real.

4.2 Dependence on combustion efficiency

The possibility of an MCE effect on the emission factor is tested by F-statistics. The emission factor for the overall data set, EF_{PN}, shows a linear relationship to MCE (R^2=0.53, F=61, $P_{err}=10^{-14}$, n=57), as does the emission factor for forest fuel (R^2=0.57, F=49, $P_{err}=10^{-11}$, n=39). Grass and savanna fuels are less conclusive, but point towards the same conclusion, i.e., a relationship between MCE and EF_{PN} (R^2=0.45, F=6, P_{err}=0.03, n=9 for savanna and R^2=0.20, F=2, P_{err}=0.24, n=9 for grass), even though it is a small data set, with a 100 nm lower size limit, and a small variation in MCE (from 0.95 to 0.98). On the other hand, there is no relationship between MCE and the CO-referenced particle emission ratios for the overall data set (R^2=0.00, F=0.3, P_{err}=0.78, n=57), while the fuel specific ratios are inconclusive (R^2=0.07, F=3, P_{err}=0.07, n=39 for forest, R^2=0.15, F=1, P_{err}=0.36, n=9 for savanna and R^2=0.34, F=4, P_{err}=0.09, n=9 for grass). We thus conclude that there is a linear relationship between EF_{PN} and MCE, but no relationship between PN/CO and MCE. The spread in the PN/CO ratios and the lack of a MCE relationship

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is shown in Fig. S1 in Supplement (http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf).

Standard fitting methods were applied to the combined data set (corrected for the 100 nm size limit) from grass, savanna, and forest fires (Eq. 6, Fig. 2), where most of the data are from flaming conditions (MCE>0.9) (Le Canut et al., 1996; Guyon et al., 2005; Kuhn et al., 2009). The fittings applied only to the forest fuel data is shown in Eqs. (S6) and (S7) in the Supplement (http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf).

$$EF_{PN}/[kg^{-1}] = (34.4 \cdot 10^{15} - 34.6 \cdot 10^{15} \times MCE) \pm 0.8 \cdot 10^{15} \quad (6)$$

5 Particle mass emissions

5.1 Literature data and average values

Particle mass emission data is frequently used in models and is more abundant than particle number emission data, but the upper particle size limit varies between the published data sets. We focus on the accumulation mode and thus, the upper particle size limit should be between the accumulation mode and the coarse mode, i.e., around one micrometer in diameter (Fuzzi et al., 2007; Reid et al., 2005).

Here data with an upper particle size limit of 1 μm, PM_1, are used together with $PM_{2.5}$ data, as the analysis of the MCE relationship then is based on 50 instead of only the 4 available PM_1 data points, and the average EF_{PM} is based on 61 instead of 11 data points (Battye and Battye, 2002; Dhammapala et al., 2007; Kaufman et al., 1992; Korontzi et al., 2003; Scholes et al., 1996; Ward et al., 1991; Ward and Hardy, 1991; Ward et al., 1992; Ward, 1996; Yokelson et al., 2007; Formenti et al., 2003; Martins et al., 1996), with the two last references excluding MCE. Data reported without relation to MCE is used only in the calculation of the arithmetic mean ± standard deviation of EF_{PM}, resulting in an EF_{PM} for the overall data of (7.6±4.7) g kg^{-1} d.m., with

larger emissions for forest fuels and smaller emissions for savanna and grass fuels (Table 2). The average PM/CO emission ratio is (0.09±0.04) g g^{-1} for the overall data, with similar fuel effects as for EF_{PM}, giving emission ratios of PM/CO=0.13±0.05) g g^{-1} for forest; PM/CO=(0.08±0.03) g g^{-1} for savanna; and PM/CO=(0.07±0.03) g g^{-1} for grass. All analyses shown here have been repeated on a dataset including also $PM_{3.5}$, PM_4 and $PM_{0.5}$ data to show the limited effect resulting from adding data with slightly different particle size limits (Tables S2 and S3 in Supplement http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf).

The EF_{PM} obtained in this study, both as direct fuel specific averages and calculated using the equations obtained below (Eqs. 7–10) with fuel specific MCE averages, are similar to other published non-parameterized PM emission factors, Table 2.

5.2 Dependence on combustion efficiency

F-statistics analysis shows that EF_{PM} is MCE dependent, while the PM/CO emission ratio has no MCE dependence (Table 3). In some studies, the CO emission factor is not given and the PM/CO emission ratio is based on the CO emission factor calculated from MCE and an estimated CO_2 emission factor of 1580 g kg^{-1} d.m. (Andreae and Merlet, 2001). The fraction of the data treated this way is between 0 and 0.42 for the different fuel types (Table 3).

Figure 3 shows the EF_{PM} vs. MCE data, with most of the available data from flaming combustion. The EF_{PM} to MCE standard linear fits for the different fuel types (Eqs. 7–9) and for the overall data set (Eq. 10) are shown as lines in Fig. 3, with the standard error of the overall data fit shown as a shaded area. The forest fire emission factors are higher than the emissions for savanna and grass fires. The overall data fit has a larger slope than the fuel specific fits possibly influenced by the high emitting – low MCE forest fuels combined with the low emitting and high MCE grass/savanna fuels. The higher EF_{PM} for forest type fuels is a combined result of both higher $EF_{PM,forest}$

(MCE) and lower MCE in the forest case.

$$EF_{PM,forest}/[g\,kg^{-1}] = (93.2 - 89.8 \times MCE) \pm 3.8 \quad (7)$$

$$EF_{PM,savanna}/[g\,kg^{-1}] = (66.8 - 65.1 \times MCE) \pm 2.5 \quad (8)$$

$$EF_{PM,grass}/[g\,kg^{-1}] = (62.9 - 62.1 \times MCE) \pm 1.1 \quad (9)$$

$$EF_{PM,overall}/[g\,kg^{-1}] = (86.1 - 85.3 \times MCE) \pm 3.1 \quad (10)$$

The standard linear fitting has been performed on some 30 published studies individually, giving similar linear fits as the ones obtained here (Table S4 in Supplement http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf). We conclude that MCE is an important variable to describe the particle mass emissions from vegetation fires.

6 Particle number emission factors derived from particle mass emission factors

The following closure study is used to determine which parameters contribute the largest uncertainty, as the available emission data on particle number, particle size distribution and particle mass is rather sparse. We compare the measurement-based fitted particle number emission factor, Eq. (6), to the particle number emission factors calculated using EF_{PM}, Eqs. (7–10), combined with different assumptions, mainly Eqs. (2) and (3) for size distribution. The particle density is assumed not to vary with MCE, and is set to 1300 $g\,m^{-3}$ (Reid et al., 2005)

6.1 Average EF_{PN}

To visualize the particle size impact on the calculated EF_{PN}, EF_{PM} is calculated for each fuel, using Eqs. (7–10) at the average MCE over all particle number emission data, MCE=0.95 (Table 1). Three different values of D_g are used (D_g=100, 130 or

150 nm, Reid et al., 2005), and the D_g to σ_g relationship for fresh particles, Eq. (3), is used. To visualize the impact from σ_g, one case using the aged data set fit, Eq. (4), is added. The effect from this change is a 20% decrease in EF_{PN}, Fig. 4.

A decrease in particle diameter D_g from 150 to 100 nm almost doubles the particle number emissions (Fig. 4), including the minor effect related to changing σ_g with D_g, Eq. (3). The particle number emission factors calculated from particle mass data compare best to the directly measured EF_{PN}, when using a diameter of ~140 nm for forest fire particles and 100 nm for grass fire particles (Fig. 4). This is in accordance with, e.g., Reid et al., 2005, where the grass/savanna fuel particles give an average D_g of 110 nm, while forest fires give 140 nm, suggesting that forest fire particles are larger than grass fire particles. This difference might be even larger taking the MCE effect on particle size into account.

6.2 MCE dependent EF_{PN}

A number of different assumptions are used to calculate the MCE dependent EF_{PN} from EF_{PM}. In Fig. 5, $EF_{PM,overall}$ is used as a basis for the calculation of EF_{PN}, and it is evident that the measured EF_{PN} is most similar to the calculated EF_{PN} when both EF_{PM} Eqs. (7–10), and the particle size distribution are allowed to vary with MCE (Eqs. 2–3). A much poorer fit is obtained when either parameter is held constant at MCE=0.95, i.e., the average combustion efficiency of the EF_{PN} data presented in Table 1. Results obtained with a wider range of assumptions are shown in the Supplement (http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf).

In Fig. 6a the fuel dependent particle mass emission factors (Eqs. 7–10) are used to calculate EF_{PN} for each fuel, while the particle size varies only with MCE and not fuel type, even though this was suggested in Fig. 5 and in Reid et al., 2005b. Keeping the size variation with MCE while introducing a fuel related size difference, we assume that grass particles are 25 nm and savanna particles are 20 nm smaller than in Eq. (2). The result is shown in Fig. 6b, where the emission factors for different fuels mainly collapse onto one line, following the results in Table 1, where no fuel effect on particle number

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emissions was found. This suggests that the fuel effect on particle mass emissions might solely result from particle size effects, with similar particle number emissions for all fires.

7 EF_{PN} estimate for coarse particles

For completeness, the particle number emission factor has also been defined for coarse particles, $EF_{PN,c}$, calculated both through the number ratio, and through the mass ratio, between the accumulation and coarse modes given in the literature. All parameters referring to the coarse mode were defined and determined in analogy with the accumulation mode parameters, but with "c" as an index. For the coarse particle emissions a relationship to MCE could not be established.

Table 4 gives a literature overview of number concentration ratios between the coarse and accumulation mode, with a median number ratio of 10^{-4}, i.e., the number of coarse particles emitted is 10 000 times smaller than the number of accumulation mode particles. If EF_{PN} for accumulation mode particles equals 10^{15} kg^{-1} d.m. (Table 1) the $EF_{PN,c}$ for coarse particles would be 10^{11} kg^{-1} d.m. The mass median ratio of $EF_{PM,c}$ and EF_{PM} from Table 4 is 0.2 ±0.1, which together with the average EF_{PM} of the overall data (7.6±4.7) g kg^{-1} d.m. gives an approximate $EF_{PM,c}$ of 1.5 g kg^{-1} d.m.

The $EF_{PM,c}$ estimate is given without fuel differences, both due to scarce available data and because a smaller effect of fuel on coarse particle emissions is assumed, based on simple calculations. We know from the literature that grass fires emit a larger proportion of coarse particles than forest fires (Reid et al., 2005; Schafer et al., 2008; Andreae and Merlet, 2001). For the ratio between EF_{PM} and $EF_{PM,c}$ we use the ratio between the emissions of total suspended particles (TSP) and $PM_{2.5}$ of 0.35 for forest and 0.54 for savanna/grassland (Andreae and Merlet, 2001), as an upper estimate compared to the other data in Table 4. Using the EF_{PM} calculated using Eqs. (7) to (9) at fuel-dependent averaged MCE (Table 2), we obtain an $EF_{PM,c}$ for the different fuels between 3 and 4 g kg^{-1} d.m. (0.54×5.1=2.8 g kg^{-1} d.m. for grass,

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$0.54 \times 6.3 = 3.1\,g\,kg^{-1}$ d.m. for savanna and $0.35 \times 11.5 = 4.0\,g\,kg^{-1}$ d.m. for forest) showing a lower effect from fuel for the coarse particles as compared to the accumulation mode particles.

To calculate the $EF_{PN,c}$ from $EF_{PM,c}$ we exemplify $EF_{PM,c}$ to be 1, 2.5, or $4\,g\,kg^{-1}$ d.m., and use different assumptions for the particle size distribution. The peak particle size is lower for number size distributions, $D_{g,c}$, than for mass size distributions, $D_{g,c,M}$ (Table 4 and Hatch-Choate equations found, e.g., in Hinds, 1998), and is exemplified with 1, 3 or 5 μm. For $\sigma_{g,c}$ 1.6, 1.8 and 2.0 are used, as 1.6 is calculated from $D_{g,c}$ and $D_{g,c,M}$ in Reid and Hobbs, 1998 (Table 4) while 2.0 describes the dust mode in the ECHAM model (Stier et al., 2005), and Haywood et al., 2003 used a $\sigma_{g,c}$ of 1.9±0.4 for the biomass coarse mode.

Table 5 shows resulting $EF_{PN,c}$ between 10^9 and $10^{12}\,kg^{-1}$ d.m., and a median value of $2 \times 10^{10}\,kg^{-1}$ d.m. The value of $EF_{PN,c} = 2 \times 10^{10}\,kg^{-1}$ d.m. for the particle mass ratio agrees fairly well with $EF_{PN,c} = 10^{11}\,kg^{-1}$ d.m. from the particle number ratio, keeping the large uncertainty in the input in mind.

8 Conclusions and outlook

We have used published data on aerosol particle number and mass emissions from vegetation fires to calculate dynamic emission factors, as a function of MCE for different fuel types. Emission factors and size distribution parameters for both accumulation and coarse mode particles are presented in relation to MCE, fuel type, and mass of dry fuel burned. While particle mass emissions, EF_{PM}, depend strongly on fuel type, we found no such relation for particle number emissions, EF_{PN}, which can be explained by differences in particle size alone.

For the emission ratio of particle number to carbon monoxide (PN/CO) we found no dependence on MCE or fuel type. The PM/CO also did not depend on MCE, but was larger for forest fires than for grass and savanna fires.

The above results make possible an efficient description of biomass burning aerosol emissions in dynamic models that provide information about MCE or CO emissions for vegetation fires, and can thus be linked directly to existing emission inventories. Models describing the climate-driven changes in fuel composition and fire evolution would, together with these dynamic emission factors, give important input to climate-related changes in vegetation fire particle number emission and CCN effects.

We must point out, however, that the parameterizations presented here are based on a very limited number of measurements and should be tested and confirmed, or refined, by further experimental studies. Well-defined laboratory experiments should help to improve the mechanistic understanding of particle emission/formation and aging, and field data are urgently needed for the validation of the above or similar parameterizations. For proper validation, the experimental studies should comprise measurements of particle number, mass and size distributions as a function of plume age and combustion efficiency.

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Table 1. Particle number emission data from experimental studies: particle size range, measurement equipment, smoke age, fuel type, modified combustion efficiency, emission factors (EF_{PN}) and emission ratios (PN/CO), only including particles larger than 100–120 nm ($PN_{>100}$, PCASP measurements) or including all particles in the accumulation mode (PN). n is the number of data points and values are tabulated as reported in the cited studies (arithmetic mean ± standard deviation when available). The Le Canut et al. data of $PN_{>100}$ were extrapolated to PN by assuming the same size distribution as reported by Formenti et al (extrapolated values in italic). Below the horizontal line the average over the three studies included in the analysis is reported, and the data in the last five lines refer to smoke outside the age range considered in this study, which are included for comparison, but not used any further.

Particle diameter (nm)	Equipment	Age	Fuel	MCE	$EF_{PN>100}$ (10^{15} kg^{-1})	EF_{PN} (10^{15} kg^{-1})	$PN_{>100}$/CO (cm^{-3} ppb^{-1})	PN/CO (cm^{-3} ppb^{-1})	n	References
3–3000	uCPC	1–30 min	Savanna	n.a.	n.a.	n.a.	n.a.	35–45	1	Hobbs et al., 2003
5–1000	CPC, PCASP	Minutes	Savanna	n.a.	n.a.	n.a.	26, 30	51, 55	2	Formenti et al., 2003
8–300	CPC	Minutes	Forest	0.94±0.02	n.a.	1.6±1.0	n.a.	27±12	34	Guyon et al., 2005
>100	PCASP	Minutes	Grass	0.96±0.01	0.66±0.32	*1.2±0.6*	19±11	36±21	9	Le Canut et al., 1996
>100	PCASP	Minutes	Savanna	0.97±0.01	0.67±0.21	*1.3±0.4*	22±10	46 ± 14	9	Le Canut et al., 1996
10–3000	CPC	Minutes	Forest	0.93±0.04	n.a.	3.4±0.6	n.a.	50±9	5	Kuhn et al., 2009
Average			All fuels	0.95±0.02	n.a.	1.7±1.2	n.a.	34±16	57	Guyon et al., 2005; Le Canut et al., 1996; Kuhn et al., 2009
> 3	uCPC	< 1 min	Savanna	0.94±0.02	n.a.	31±19	n.a.	550±310	4	Sinha et al., 2003
>3	PCASP	<1 min	Savanna	0.95±0.02	0.21±0.15	n.a.	4.8±4.1	n.a.	8	Sinha et al., 2003
100–3000	PCASP	days	Grass	0.98	0.4	n.a.	23	n.a.	2	Anderson et al., 1996
120–3000	PCASP	10 days	n.a.	n.a.	n.a.	n.a.	n.a.	12.2^{RH}	1	Andreae et al., 1994
>300	LIDAR	days	n.a.	0.97	n.a.	n.a.	n.a.	17	1	Browell et al., 1996

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Table 2. Particle mass emission factors, EFPM, in g kg^{-1} d.m. (arithmetic mean ± standard deviation) for the three fuel categories separately and the overall data set; the number of data points in the average (n); EFPM calculated using the mean MCE for each fuel subset, in the fuel specific fitted equations (Eqs. 7 to 9); the number of data points in the fits (n_{fit}) compared to previous emission factor reviews.

Fuel	EF_{PM}, Average [g kg^{-1}]	n	EF_{PM} ($MCE_{fuel\ type}$) [g kg^{-1}]	n_{fit}	EF_{PM}, Reid et al., 2005 [g kg^{-1}]	EF_{PM}, Andreae and Merlet, 2001 [g kg^{-1}]
Forest	9.6±4.6	21	11.5±4.5	12	15±11[b]	10±3[d]
Savanna	6.3±3.0	24	6.3±2.0	24	8±2[b]	
Grass	4.7±2.1	15	5.1±1.9	14	7±2[b]	5± 2
Overall data	7.6±4.7	61	7.6±3.4	50		7±2[c]

[a] Averaged over the available data, not taking burned amount into consideration. [b] Given with "absolute uncertainty" instead of standard deviation. [c] Average weighted with burned mass (Andreae and Merlet, 2001). [d] Average over two different kinds of forests, weighted with burned mass (Andreae and Merlet, 2001).

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Table 3. Emission ratio (PM/CO) and emission factor for particle mass (EF_{PM}) for the overall data set and the fuel specific data subsets as an arithmetic mean ± standard deviation. Both variables presented with the correlation coefficient (R^2); the F-statistic (F); and the probability that the F-statistic erroneously shows a relation (P_{err}). CO_{calc} is the fraction of the data where CO emissions were not reported and thus calculated as described in the text; MCE is the modified combustion efficiency averaged over each fuel subset and the overall data, n is the number of data points used in the analysis and n_s the number of published studies used.

Fuel	EF_{PM} [g kg^{-1}]	R^2	F	P_{err}	PM/CO [g g^{-1}]	R^2	F	P_{err}	CO_{calc}	MCE	n	n_s
Forest	11±6	0.60	15	10^{-3}	0.13±0.05	0.27	4	0.06	0.42	0.91±0.05	12	4
Savanna	6±3	0.33	11	410^{-4}	0.08±0.03	0.05	1	0.37	0	0.93±0.03	24	4
Grass	5±2	0.74	34	10^{-5}	0.07±0.03	0.15	2	0.16	0.13	0.93±0.03	14	3
Overall data	7±4	0.48	44	210^{-11}	0.09±0.04	0.05	3	0.08	0.14	0.92±0.04	50	9

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Table 4. The ratio of coarse to accumulation mode particle concentrations from different studies is given both as number ratios and mass ratios (PN_c/PN; PM_c/PM), together with the approximate peak particle size for the coarse particle mode for number size distributions ($D_{g,c}$) and for mass size distributions ($D_{g,c,M}$), where available. The fuel, or site of the burn is given in the last column and the median of the data in the last row.

Cite	PN_c/PN	PM_c/PM	$D_{g,c}$ [μm]	$D_{g,c,M}$ [μm]	Fuel or site etc.
Andreae et al., 1994	2.00E–05				savanna, forest
Hungershoefer et al., 2008	1.00E–04		4		lab, grass
Hungershoefer et al., 2008	1.00E–04		2–3		lab, musasa
Haywood et al., 2003	1.00E–04		3		Otavi plume
Radke et al., 1991	1.00E–06		<10		boreal forest
Petzold et al., 2007	2.00E–04		1–2		very old, boreal forest
Le Canut et al., 1996	1.00E–04		>3		savanna, grass
Reid and Hobbs, 1998	2.00E–05	0.1	1.5	3	Brazil, all fuels
Keshtkar and Ashbaugh, 2007		0.15		10	lab, agriculture
Fuzzi et al., 2007		0.2		4–5	Brazil
Fuzzi et al., 2007		0.1		5	Brazil
Eck et al., 2003		0.14		1.5	aged, peat and forest
Eck et al., 2003		0.1		5–10	forest
Eck et al., 2003		0.3		7	grass
Andreae and Merlet, 2001		0.54			grass/savanna
Andreae and Merlet, 2001		0.35			forest
Ward et al., 2006		0.3			grass/savanna
Median	1.00E–04	0.2			

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Table 5. The different $EF_{PN,c}$ calculated with three different assumptions about the ratio between particle mass concentrations in the coarse and accumulation modes (~10%, ~30% and ~50% of EF_{PM}), three different $D_{g,c}$ (1, 3, or 5 μm) and three different $\sigma_{g,c}$ (1.6, 1.8, or 2.0). The $D_{g,c,M}$ are calculated from $D_{g,c}$ and $\sigma_{g,c}$ using the Hatch-Choate equations (e.g., Hinds, 1998). All cases use $EF_{PM,overall}$=7.6±3.4 g kg^{-1} d.m. for the accumulation mode particle mass emission factor for the overall data set.

$D_{g,c}$ [μm]	$\sigma_{g,c}$	$D_{g,c,M}$ [μm]	$EF_{PN,c}$ [10^9 kg^{-1}]		
			$EF_{PM,c}$=1 g kg^{-1}	$EF_{PM,c}$=2.5 g kg^{-1}	$EF_{PM,c}$=4 g kg^{-1}
1	1.6	2	5.4E+02	1.4E+03	2.2E+03
1	1.8	3	3.1E+02	7.8E+02	1.2E+03
1	2.0	4	1.7E+02	4.2E+02	6.8E+02
3	1.6	6	2.0E+01	5.0E+01	8.1E+01
3	1.8	8	1.2E+01	2.9E+01	4.6E+01
3	2.0	13	6.3E+00	1.6E+01	2.5E+01
5	1.6	10	4.4E+00	1.1E+01	1.7E+01
5	1.8	14	2.5E+00	6.2E+00	9.9E+00
5	2.0	21	1.4E+00	3.4E+00	5.4E+00

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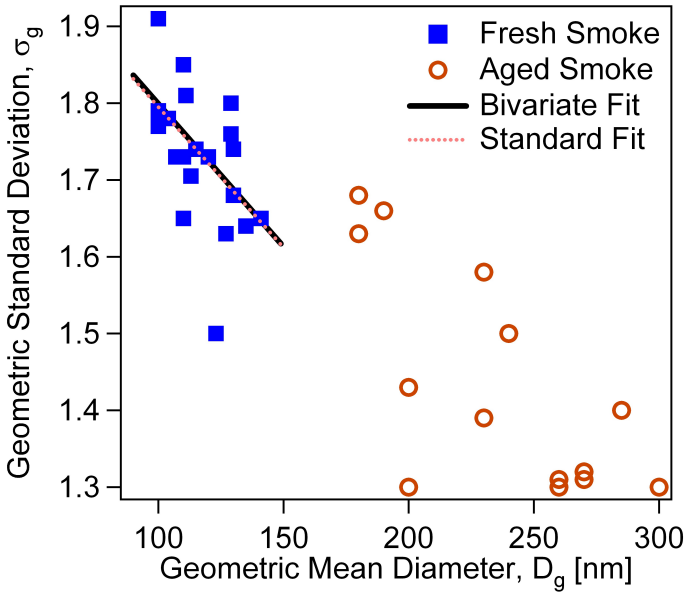

Fig. 1. The geometric mean diameter (D_g) versus the geometric standard deviation (σ_g) for published fresh and aged smoke data. A bivariate linear fitting method (Cantrell, 2008) has been used on the fresh smoke data, yielding D_g/[nm]=(584±5)–(269±1)×σ_g, Eq. (3), shown as a line. The dotted line is found by fitting the fresh smoke data with the standard regression method. All data are listed in the Supplement (http://www.atmos-chem-phys-discuss.net/9/17183/2009/acpd-9-17183-2009-supplement.pdf).

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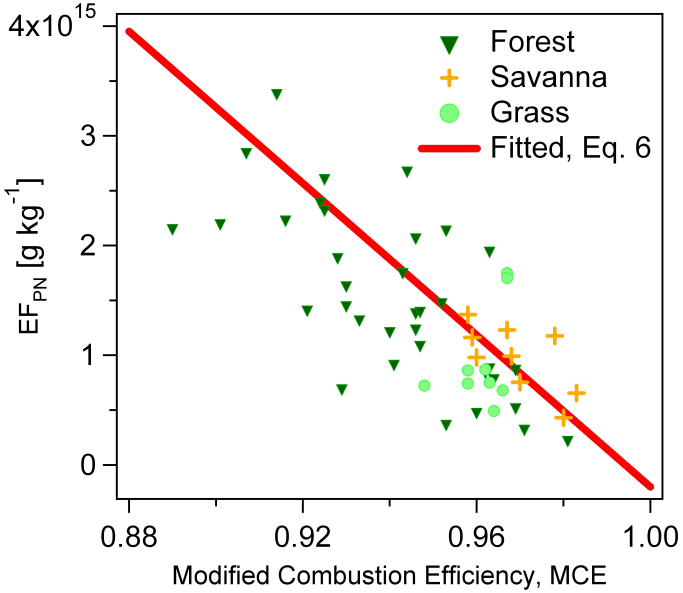

Fig. 2. Particle number emission factors (EF_{PN}) related to dry mass burned versus modified combustion efficiency (MCE) for three fuel types; forest (Guyon et al., 2005; Kuhn et al., 2009), and savanna and grass (Le Canut et al., 1996). A standard fitting method is used on the overall data set to find $EF_{PN}/[kg^{-1}]=(34.4 10^{15}–34.6 10^{15} \times MCE) \pm 0.8 10^{15}$, Eq. (6), shown as a line. Measurements for savanna and grass data used a ~100 nm particle diameter detection limit and have been corrected as described in Sect. 4.1.

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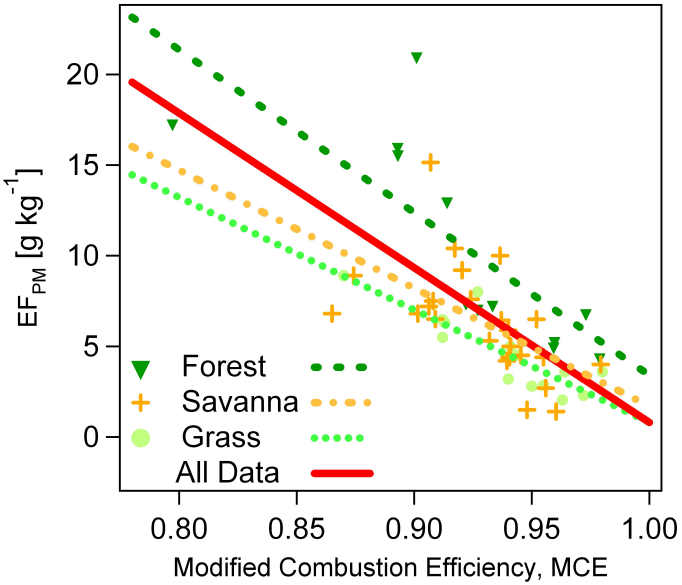

Fig. 3. Particle mass emission factors (EF_{PM}) related to dry mass burned versus modified combustion efficiency (MCE) for three fuel types from ten different studies, with the fitted equations for each of the fuel types and for the overall data set ($EF_{PM,forest}/[g\,kg^{-1}]=(93.2–89.8\times MCE)\pm3.8$, for forest, $EF_{PM,savanna}/[g\,kg^{-1}]=(66.8–65.1\times MCE)\pm2.5$ for savanna, $EF_{PM,grass}/[g\,kg^{-1}]=(62.9–62.1\times MCE)\pm1.1$, for grass and $EF_{PM,overall}/[g\,kg^{-1}]=(86.1–85.3\times MCE)\pm3.1$ for the overall data set, Eqs. 7–10) shown as lines.

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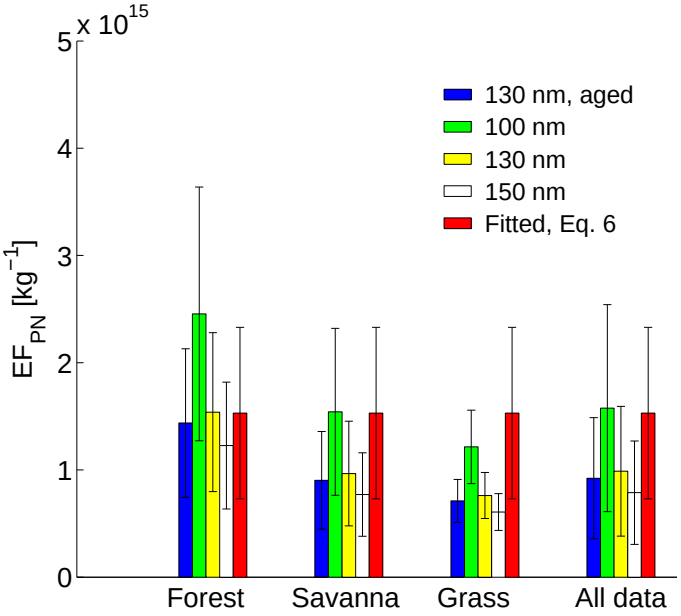

Fig. 4. Particle number emission factors (EF_{PN}) related to dry mass burned, calculated from EF_{PM} at MCE=0.95 for the different fuel types, Eqs. (7–10), using different particle diameters (D_g) given in the legend. The geometric mean diameter D_g is related to the geometric standard deviation σ_g using Eq. (3), apart from the "130 nm, aged" case, where the fit to the aged data set, Eq. (4), is used. The calculated EF_{PN} for different particle sizes are compared to the measurement-based fitted equation, $EF_{PN}/[kg^{-1}]=(34.4 10^{15}-34.6 10^{15}\times MCE)\pm 0.8 10^{15}$, Eq. (6), at MCE=0.95. The error bars are based on the standard error of the fits for EF_{PM} vs MCE and EF_{PN} vs. MCE respectively.

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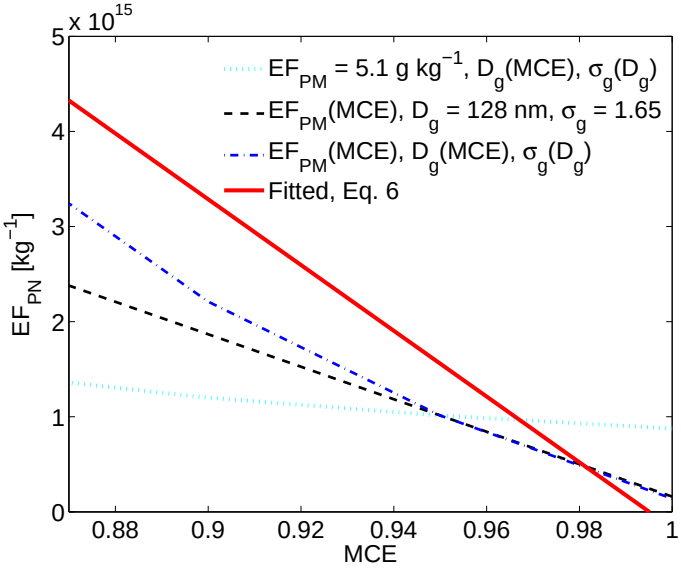

Fig. 5. Particle number emission factors (EF_{PN}) related to dry mass burned versus modified combustion efficiency (MCE), calculated from the particle mass emission factor for the overall data set ($EF_{PM,overall}$). $EF_{PM,overall}$ is either constant at MCE=0,95; $EF_{PM,overall}$=5.1 g kg^{-1} d.m., or varied with MCE, Eq. (10). The particle sizes are either constant at MCE=0.95; D_g=128 nm and σ_g=1.65, or varied with MCE, Eqs. (2–3).

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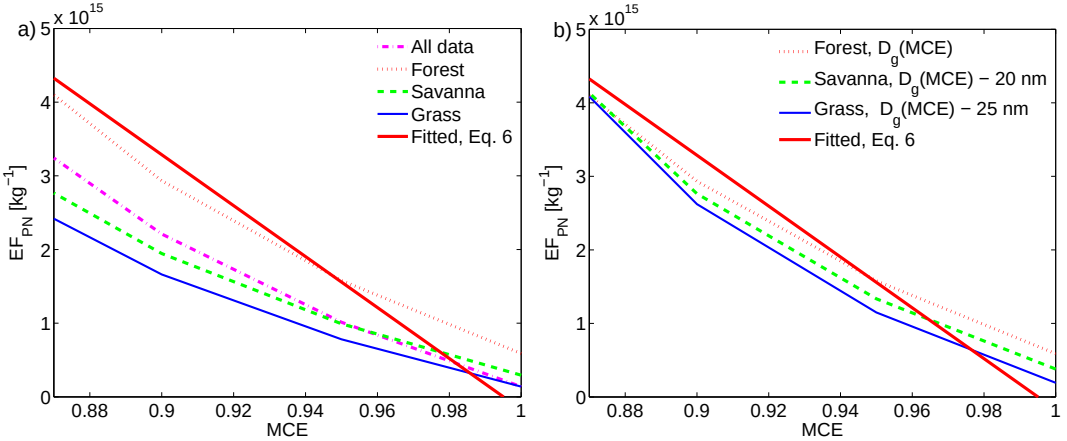

Fig. 6. Particle number emission factors (EF_{PN}) related to dry mass burned versus modified combustion efficiency (MCE) for different fuels. The calculations from EF_{PM} data are based on varying EF_{PM} with fuel and MCE, Eqs. (7–10), and the particle sizes with MCE Eqs. (2–3). **(a)** No particle size difference between fuels is assumed. **(b)** The particle size has been reduced for grass emissions by 25 nm and for savanna emissions by 20 nm, in accordance with Reid et al., 2005.

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