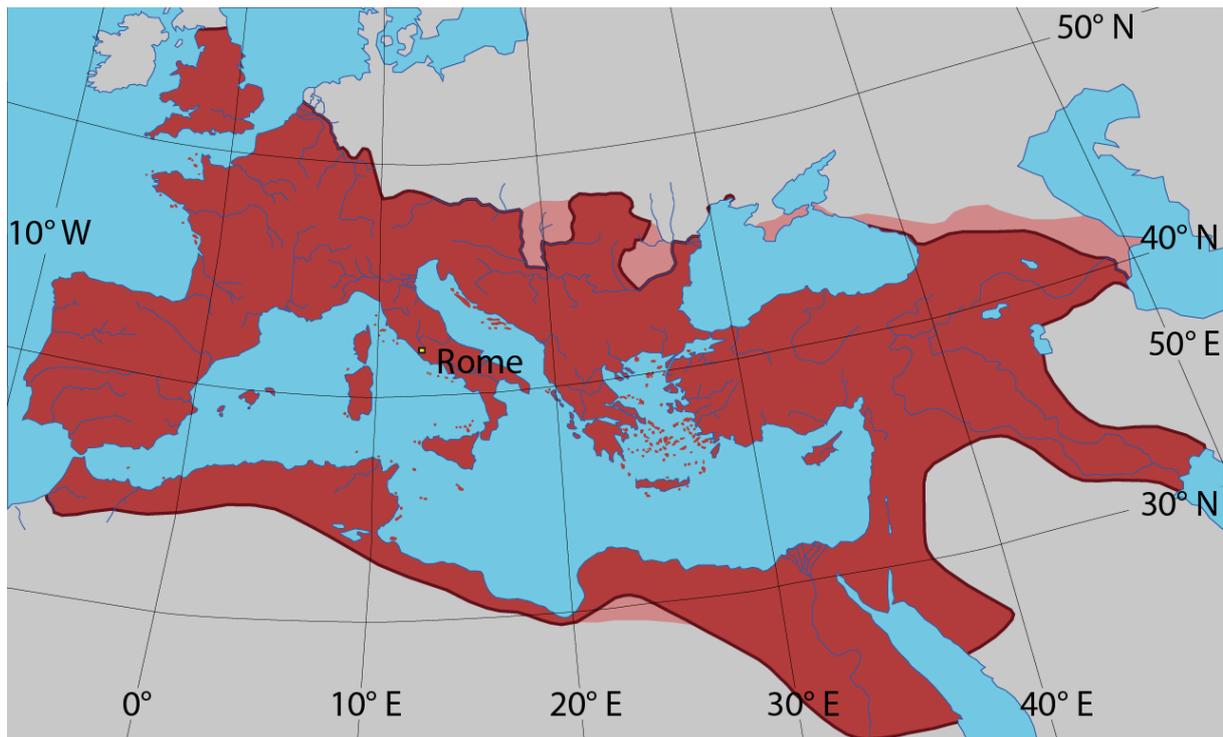


## S1 Map of the Roman Empire



**Fig. S1:** The Roman Empire at its greatest extent (AD 117) with its vassals in pink. Adapted from Tataryn (2016); licensed under the Creative Commons Attribution-Share Alike 3.0 Unported license (<https://creativecommons.org/licenses/by-sa/3.0/deed.en>).

## S2 Model ~~set-up~~setup

~~Illustrated are the setups of the 6 simulations conducted with ECHAM-HAM-SALSA (no\_humans, LCC\_HYDE, LCC\_KK, LCC\_HYDE\_low, LCC\_HYDE\_int, and LCC\_KK\_high). Models that we used are shown in dark colours, whereas inputs to these models are shown in light colours. ECHAM-HAM-SALSA (violet) includes (among other components) the vegetation model JSBACH, a secondary organic aerosol scheme, and a sulfur cycle. Natural fire emissions were calculated with CBALONE-SPITFIRE (orange). For driving the two models, output from the Earth System Model MPI-ESM was used among others.~~

**Tab. S1:** Greenhouse gas concentrations and orbital parameters. Precession is expressed as the longitude of the perihelion with respect to the equinox. The values are averages over AD 50 to 150.

Var.	Unit	Value
CO <sub>2</sub>	ppm	278
CH <sub>4</sub>	ppb	662
N <sub>2</sub> O	ppb	267
Eccentricity	-	0.01742
Precession	<del>degree</del> <u>degrees</u>	250.8
Obliquity	<del>degree</del> <u>degrees</u>	23.68

**Tab. S2:** Overview of ~~some~~natural fire aerosol, anthropogenic aerosol, and SOA precursor emissions in the different ECHAM-HAM-SALSA simulations. The multiplication factor refers to the natural fire emissions calculated with CBALONE-SPITFIRE.  $l$  stands for the fraction of the vegetated area that is covered by anthropogenic land (crop and pasture) and AA for anthropogenic aerosol emissions.

Simulation	Multiplication factor	AA	SOA precursor emissions
no_human	1	No	based only on natural land cover
LCC_HYDE	1	No	changed due to anthropogenic land cover
LCC_KK	1	No	changed due to anthropogenic land cover
LCC_HYDE_low	$(1 - l)$	Yes	changed due to anthropogenic land cover
LCC_HYDE_int	$(1 - l)$	Yes	changed due to anthropogenic land cover
LCC_KK_high	$(1 - l)$	Yes	changed due to anthropogenic land cover

### S3 Aerosol emissions from fires

CBALONE-SPITFIRE needs forcing data related to the vegetation and the atmosphere at a daily time resolution as well as a starting point for the carbon pools. To drive CBALONE-SPITFIRE we used data from MPI\_no\_LCC (illustrated in Fig. ??) from which output at a high temporal resolution was saved for some selected 30-year periods. The driving data for the AD 100 CBALONE-SPITFIRE simulations represents a slightly earlier period (around AD 1). However, we do not expect a significant difference between AD 1 and AD 100 since the forcing in MPI\_no\_LCC is very similar for the two years. The 30-year period around AD 1 was repeatedly used to drive both the spin-up ( $\approx 100$ -years) and the analysed simulated period of CBALONE-SPITFIRE. In the following, we will refer to these fire emissions that are technically for AD 1 as those of AD 100 for simplicity. The emissions have a daily resolution and show interannual variability.

We also calculated fire emissions around AD 1835 (not shown). These emissions were not used as input for our ECHAM-HAM-SALSA simulations listed in Table S2 but for comparison with the fire emissions from van Marle et al. (2017, Sect. S9). We again used high time resolution output from an MPI-ESM simulation to drive CBALONE-SPITFIRE, called MPI\_LCC in the following. The only difference between MPI\_no\_LCC and MPI\_LCC is that the latter considers anthropogenic land cover change.

Next to carbon pools and variables related to the vegetation or the atmosphere, the fire model depends on the following additional inputs: i) lightning; ii) population density, together with a regionally varying anthropogenic influence factor  $a_n$ .

To estimate the lightning frequency, we tested the parameterisation by Magi (2015) that is based on convective precipitation fluxes. We found that the such-derived lightning frequency is very similar for AD 100 and AD 1835. However, the simulated lightning frequency differs from the standard lightning frequency used by the MPI-ESM implementation of SPITFIRE (e.g. different spatial pattern; not shown), which is based on the Lightning Imaging Sensor/Optical Transient Detector (LIS/OTD) as described in Lasslop et al. (2014). The difference between the simulated and observation-based lightning frequency is probably associated with the high uncertainties in the parameterised convection scheme. Furthermore, the parameterisation by Magi (2015) works better for convective mass flux through the 0.44 hybrid-sigma pressure level than for convective precipitation, but the former was not available from the MPI-ESM simulations. Thus, we decided to use the observationally derived lightning frequencies by LIS/OTD for both AD 100 and AD 1835 (Fig. ??) in order to not introduce a large bias in natural fire ignitions.

Humans impact fires directly by ignitions and fire fighting as well as indirectly, e.g. by forest management and landscape fragmentation. It is very likely that the relationship between humans and fires has changed in the past centuries and millennia as a consequence of large cultural and political changes. This is neglected in SPITFIRE since  $a_n$  does not change over time. Instead,  $a_n$  (and thus the number of anthropogenic ignitions) in SPITFIRE is tuned towards present-day fires where satellite data is available.

For estimating the fire emissions in AD 1835, we nevertheless used CBALONE-SPITFIRE with the present-day dependence on population density (i.e. the standard  $a_n$ ), as it was done by the FIREMIP project for calculating fire emissions from 1750 to today. For these CBALONE-SPITFIRE simulations, we averaged the population density from HYDE11 over 1830 and 1840. Furthermore, we considered anthropogenic land cover change when calculating the fire emissions; the same changes were used as in MPI\_LCC.

For AD 100, we used CBALONE-SPITFIRE to calculate natural fire emissions (Fig. ??);

~~assuming no anthropogenic influence. Hence, no anthropogenic land cover change was considered in these CBALONE-SPITFIRE simulations. Furthermore, the population density was set to 0 and  $a_n$  thus becomes irrelevant. The ECHAM-HAM-SALSA simulations LCC\_HYDE\_low, LCC\_HYDE\_int, and LCC\_KK\_high account for anthropogenic aerosol emissions, which include agricultural burning. Using the same natural fire emissions in these simulations than in the other simulations (no\_human, LCC\_HYDE, and LCC\_KK) would lead to an overestimation in total aerosol emissions since natural aerosol emissions should not occur where now crop or pasture grows. Therefore, we reduced the natural fire emissions of the simulations LCC\_HYDE\_low, LCC\_HYDE\_int, and LCC\_KK\_high offline to account for regions subject to anthropogenic land use (Table S2; Fig. ??). As a first order approximation, the natural fires calculated with CBALONE-SPITFIRE were multiplied with  $(1-l)$ , where  $l$  is the fraction of the vegetated area per gridbox that is covered by crop and pasture.~~

### S3 Aerosol emission factors for fuel consumption

The fuel consumption studies that we considered mostly provide emission factors for either open fire places or traditional stoves. An exception is the measurement of olive pits that we included, which was conducted in a pellet stove; it was the only measurement of olive pits that we could find, and olive oil pressing waste was employed as a domestic and industrial fuel throughout Antiquity in the Mediterranean (Rowan, 2015).

#### S3.1 Wood

For wood combustion, we considered many wood types which were abundant in the Roman Empire (e.g. different types of oak, olive tree, maritime pine, or beech). However, many of these measurements were conducted with the same burning devices (Alves et al., 2011; Calvo et al., 2015; Fernandes et al., 2011). Looking at different studies, we observed that the differences in  $EF$ s between different wood types are sometimes smaller than the differences between different measurement setups. Hence, we also decided to include some trees in our compilation which were not present in the Mediterranean region (e.g. Lespedeza, Paulownia) to consider more independent studies. The medians and quartiles that were derived for the emission factors of wood ( $EF_{\text{wood}}$ ) from the random sampling can be found in Table S3.

#### S3.2 Agricultural waste

For agricultural waste, a large variety of burning material was considered (e.g. wheat straw, rice straw, dung). Although the composition of agricultural residues in the Roman Empire differs from our compilation (e.g. rather *Triticum dicoccum* and barley instead of *Triticum aestivum* and rice; olive pits and chickpeas instead of peanuts and soybean residues), we assume that the emission factors are not fundamentally different: our compilation considers a large range of agricultural waste types. Furthermore, based on our compiled data, the measurement setup seems to be as important as the specific fuel type. The calculated medians and quartiles for the emission factors of agricultural waste ( $EF_{\text{agri}}$ ) can be found in Table S4.

#### S3.3 Charcoal

Compared to wood and agricultural waste, only few studies estimated aerosol  $EF$ s for charcoal burning and charcoal making. Furthermore, most of them provide measurements of total suspended particles (e.g. Smith, 2000), and not for BC and OC. Thus, we considered only the very

recent study by Keita et al. (2018) for BC and OC, who measured BC and OC for charcoal cooking fires and charcoal making in West Africa and calculated emission factors. Maybe even more important than knowing the exact BC and OC emission factors for charcoal burning is considering also the emissions from charcoal making. To produce 1 kg of charcoal, approximately 7 kg of wood are needed with traditional methods (Olson, 1991). As a consequence, using charcoal instead of wood as fuel might overall result in similar or even higher aerosol emissions, since for every kilogramme of charcoal burnt, the aerosols emitted during the production of this kilogramme of charcoal should also be considered. The aerosol emission factors per kilogramme of wood used for charcoal ( $EF_{ch_w}$ ) were therefore calculated as:

$$EF_{ch_w} = \frac{1}{7} \cdot EF_{ch_b} + EF_{ch_m}, \quad (1)$$

where  $EF_{ch_b}$  is the emission factor for charcoal burning (per kilogramme of charcoal) and  $EF_{ch_m}$  is the emission factor for charcoal making (per kilogramme of wood). The emission factors for  $EF_{ch_b}$  and  $EF_{ch_m}$  can be found in Tables S5 and S6, respectively.

Considering that charcoal is often cited as almost smokeless (e.g. Wood and Baldwin, 1985; Lohri et al., 2016), the measured  $EF_{ch_b}$  of BC (median:  $0.59 \text{ g kg}^{-1}$ ; comparable to the burning of other types of biofuel) is relatively high – a discrepancy that already Bond et al. (2004) noted for the emission factors of total suspended particles. In the future, more measurements could help to better understand this inconsistency.

Inserting the estimates of  $EF_{ch_b}$  and  $EF_{ch_m}$  in the equation above results in  $EF_{ch_w}$  of  $0.26 \text{ g kg}_{\text{wood}}^{-1}$  (0.18, 0.39) for BC,  $3.94 \text{ g kg}_{\text{wood}}^{-1}$  (3.27, 4.80) for OC, and  $0.21 \text{ g kg}_{\text{wood}}^{-1}$  (0.14, 0.32) for  $\text{SO}_2$ .

### S3.4 Combining different sectors

To assess the overall emission factor, we needed to estimate how much which sector contributed to the total fuel consumption. Except for  $\text{SO}_2$ , the emission factors are similar for wood and agricultural waste. The OC emission factors for charcoal burning and production (expressed as per kilogramme wood) are larger than for wood and agricultural waste, whereas the opposite is the case for BC. Thus, different assumptions concerning the contributions from the three sectors would affect the BC to OC ratio, rather than the ~~overall~~ emissions of both of them.

Wood fuel (including wood for charcoal making) was the dominant fuel in the Roman Empire (Olson, 1991). However, agricultural residues such as chaff, olive pits, and dung were also used, most evidently in regions lacking in supplies of wood (e.g. Roman North Africa and Roman East; Mietz, 2016; Rowan, 2015). In developing countries in 1985, the mass contribution of agricultural waste to total biofuel combustion (excluding burning in fields) ranges from 14% in Africa to over 40% in Asia (Yevich and Logan, 2003). Based on these numbers, we assumed that 20% of the used fuel consisted of agricultural waste.

Like for developing countries (Wood and Baldwin, 1985; Yevich and Logan, 2003; Lohri et al., 2016), the use of charcoal was especially important in urban areas in ancient times (Veal, 2017). Veal (2017) assumes that in the cities “perhaps 80%” of the burnt fuel consisted of charcoal with the remainder being wood, whereas the opposite ratio occurred in rural areas. In her two extreme case scenarios for Rome, she used charcoal contributions of 80% and 20% to total fuel. Assuming a conversion factor of 7, this means that 97% and 64% of the wood fuel was used for charcoal making, respectively.

These estimates are higher than present-day estimates in countries where charcoal is pro-

duced: using data from the food and agricultural organisation (FAO<sup>1</sup>), we calculated the contribution of charcoal production to total wood fuel production. We chose the year 1970, when fossil fuels were likely (even) less common in developing countries than today. A factor of 6 was used<sup>2</sup> to convert the weight of charcoal (metric tonnes) to the volume of wood (m<sup>3</sup>) required to make the specified charcoal weight. It is unclear whether countries include the amount of wood used for charcoal making in the woodfuel statistics which they report to FAO<sup>3</sup>. To account for the fact that the data might be inconsistent, we used two methods (DIV1 and DIV2) to calculate the contribution of charcoal to wood fuel ( $frac_{DIV1}$  and  $frac_{DIV2}$ ):

$$frac_{DIV1} = \frac{Wood_{ch}}{Wood_{con} + Wood_{noncon}} \quad (2)$$

and

$$frac_{DIV2} = \frac{Wood_{ch}}{Wood_{ch} + Wood_{con} + Wood_{noncon}}, \quad (3)$$

where  $Wood_{ch}$  stands for the production of wood used for charcoal making,  $Wood_{con}$  for the production of coniferous fuel wood, and  $Wood_{noncon}$  for the production of non-coniferous fuel wood. With method DIV1, we arrive at values above 100% for some countries. Since this makes no sense, we cap these values at 100%.

We calculated the mean and the median fractions for both the world and Africa, the latter having the largest per capita use of charcoal in the developing world (Yevich and Logan, 2003). For both cases, we excluded all countries that do not produce charcoal, i.e. mainly developed countries that predominantly use fossil fuels. The values are shown in Table S7. The values lie in a relatively narrow range between 13% and 25%. Although the large majority of people living in developing countries mainly used biomass for domestic energy (Wood and Baldwin, 1985), it is possible that the use of fossil fuels might affect our estimates to some degree.

Overall, the FAO data shows lower contributions of charcoal to total fuel wood than the estimates by Veal (2017). This could indicate that the estimates by Veal (2017) are too high for the whole Roman Empire (which was not the target of her study), since the Empire consisted of many parts that differed considerably with respect to wood supply. However, the Roman Empire had a high urbanisation rate ( $\approx 10\%$ , with higher values in Italy; Temin, 2006) and metallurgy was important (Harris, 2013), which speaks for a potentially higher contribution of charcoal than under present-day conditions. In the end, we decided to use a fraction of 50% of charcoal wood to total fuel wood, which is a compromise between the estimates by Veal (2017) and the FAO based values. To summarise, we assume that 20% of the fuel consisted of agricultural waste, 40% of charcoal (in terms of wood needed for charcoal production), and 40% of wood.

**Tab. S3:** Aerosol emission factors for wood burning ( $EF_{wood}$ ) in  $g\ kg^{-1}$  used for the low, the intermediate, and the high scenarios.

	Low estimate	Intermediate estimate	High estimate
BC	0.27	0.42	0.65
OC	0.84	2.09	4.15
SO <sub>2</sub>	0.0021	0.098	0.20

<sup>1</sup> <http://www.fao.org/faostat/en/#data/F0>, downloaded: 18 August 2018

<sup>2</sup> <http://www.fao.org/docrep/005/y4450e/y4450e13.htm>, last access: 27 November 2018; a similar value of 7 is mentioned here: <http://www.fao.org/docrep/x5667e/x5667e04.htm>, last access: 17 Januar 2019

<sup>3</sup> <http://www.fao.org/docrep/005/y4450e/y4450e13.htm>

**Tab. S4:** Aerosol emission factors for agricultural waste burning (as fuel;  $EF_{agri}$ ) in  $\text{g kg}^{-1}$  used for the low, the intermediate, and the high scenarios.

	Low estimate	Intermediate estimate	High estimate
BC	0.25	0.45	0.85
OC	1.16	2.40	4.32
SO <sub>2</sub>	0.0082	0.022	0.050

**Tab. S5:** Aerosol emission factors for charcoal burning ( $EF_{chb}$ ) in  $\text{g kg}^{-1}$  (per kg charcoal) used for the low, the intermediate, and the high scenarios.

	Low estimate	Intermediate estimate	High estimate
BC	0.44	0.59	0.79
OC	0.45	0.96	2.03
SO <sub>2</sub>	0.29	0.43	0.62

**Tab. S6:** Aerosol emission factors for charcoal production ( $EF_{chm}$ ) in  $\text{g kg}^{-1}$  (per kg wood) used for the low, the intermediate, and the high scenarios.

	Low estimate	Intermediate estimate	High estimate
BC	0.11	0.18	0.28
OC	3.21	3.81	4.51
SO <sub>2</sub>	0.10	0.15	0.23

**Tab. S7:** The percentages of wood used for charcoal making to total fuel wood. Two different methods (DIV1 and DIV2) were applied. Data was taken from FAO (year 1970).

Method	World, mean	World, median	Africa, mean	Africa, median
$frac_{DIV1}$	25%	16%	24%	16%
$frac_{DIV2}$	18%	13%	18%	14%

## S4 Estimating crop yield $Y$

The agronomist Columella stated that the seed-yield ratios for most parts of Italy were seldom above 4:1 in the first century AD (Spurr, 1986). Assuming a sowing amount of  $135 \text{ kg ha}^{-1}$  (advocated by the agronomists as a typical sowing amount; Goodchild, 2007), a ratio of 4:1 results in yields of  $540 \text{ kg ha}^{-1}$ . This yield is quite low compared to other sources, which show that yields of 5:1 and 6:1 were the most frequent in Italy and higher yields of 10-15:1 were not infrequent (Goodchild, 2007). For some regions, Varro (who lived in the first century BC) and Pliny the Elder (first century AD) reported even yields of 100:1 (Sinclair, 1998). So far, the estimates referred to Italy alone, but the variability between different Mediterranean countries was of course large: around AD 1921-1930 the average yields according to Hopkins (2017) ranged from values below  $600 \text{ kg ha}^{-1}$  (Cyrenaica, Tunisia, Algeria), through values between 600 and  $1000 \text{ kg ha}^{-1}$  (Greece, Portugal, Cyprus, Syria and Lebanon, Turkey, Spain) up to values above  $1000 \text{ kg ha}^{-1}$  (Bulgaria, Yugoslavia, Italy, France, Egypt). Overall, we concluded that yields representative for the whole Roman Empire roughly lie in the range between  $500 \text{ kg ha}^{-1}$  and  $1000 \text{ kg ha}^{-1}$ .

## S5 Estimating

~~There are several options for what happened after harvest with the remaining residue on the field: when chaff was short, straw could be used as livestock feed or the residues left could be~~

directly grazed by ruminants, as it happens also nowadays e.g. in Mediterranean North Africa. However, Spurr (1986) states that animals rarely eat stubble but rather the edible weeds and grass which grow among it. Farmers that had no further use for the stubble left after harvesting burnt it. In ancient sources, burning was mentioned as method to destroy weeds or to fertilise the soil. Spurr (1986) assumed that especially small farmers who could not store straw and had few animals to feed would burn the stubble.

Nowadays, crop residue burning is not widespread in developed countries anymore, and many countries in western Europe even forbid open field burning. To have a rough indication for how much of the remaining residue might have been burnt in the Roman Empire, we used present-day estimates from developing countries as an indication (also countries that cultivate rice despite the fact that rice was not grown in the Roman Empire). Yevich and Logan (2003) found that in 1985 of the available residue, 1%, 23%, and 38% was burnt in the fields in China, in India, and in the rest of Asia, respectively. Note that a more recent estimate for crop residue burning in China arrives at much higher fractions than the study by Yevich and Logan (2003): based on satellite data, Li et al. (2016a) find that 23% of the field area is burnt. A large part of the discrepancy can be explained by the different year: Yevich and Logan (2003) derive estimates for 1985, whereas Li et al. (2016a) analyse the year 2012. Field surveys from China show that the proportion of residue that is burnt is larger when the crops are harvested by a combine harvester compared to manual harvesting. In line with this, Li et al. (2016b) estimate that the fractions of crop residues burnt in fields increased from 5% in 1990 to 23% in 2013. Another reason why crop residue burning has increased might be that the use of biofuels has decreased. Note that we do not expect that mechanisation leads to enhanced crop residue burning in general; modern-day technology can also be used to prepare fields for the next crop planting after harvest, which makes the burning of crop residue unnecessary.

In the Philippines and Indonesia, up to 65% and 73% of the residue was burnt in fields in 1985. This is similar to the satellite-derived fraction of rice fields under burning for the Punjab region in India in 2015; as reasons for the large burning in the Punjab region, highly mechanised farming, poor storage facility for the straw, and lack of market demand for further use are mentioned among others. A survey in Bangladesh shows that only 3% of the farmers burn the total residue but that 37% burn the lower part of it; the surveyed farmers manually gather the residue from the field.

By the use of  $p_b$ , we have already accounted for the part of residue used as biofuel. If we remove this part of the residue from the estimates above, then the fractions of burning in the field would increase. Based on all the studies that we have mentioned, we estimate that 40% of the crop area is annually burnt for the simulation LCC\_HYDE\_int ( $Fr_{\text{crop burnt}} = 0.4 \text{ y}^{-1}$ ). For the low and the high scenarios, we changed this fraction by a factor of 2 and arrive at 20% and 80%, respectively. The 80% are reached under present-day conditions in the countries with most crop residue burning (e.g. Indonesia). Pfeiffer et al. (2013) use a value of 20% in their pre-industrial fire model (LPJ-LMfire v1.0) and consider this to be a conservative estimate.

## S5 Estimating $F$

Values of fuel loads ( $\text{kg}_{\text{dry matter}} \text{ per area}$ ) frequently lie in the range between 0.3 and 0.5  $\text{kg m}^{-2}$  for pastures and grasslands: for Kentucky Bluegrass, 0.26, 0.36, and 0.64  $\text{kg m}^{-2}$  were measured in Idaho; values for grasslands in Australia are around 0.32  $\text{kg m}^{-2}$  and 0.46  $\text{kg m}^{-2}$ ; in South Africa, total aboveground fuel loads in savanna parklands range from 0.22 to 0.55  $\text{kg m}^{-2}$ , whereas the fuel loads of standing herbaceous material range from 0.30 to 0.41  $\text{kg m}^{-2}$ ; a total

fuel load of  $0.49 \text{ kg m}^{-2}$  for Mediterranean grasslands in Greece was measured.

Fuel biomass consumptions for savannas are comparable:  $0.26 \text{ kg m}^{-2}$  for savanna woodlands for early dry season burns,  $0.46 \text{ kg m}^{-2}$  for savanna woodlands for mid/late dry season burns,  $0.21 \text{ kg m}^{-2}$  for savanna grasslands for early dry season burns, and  $1.0 \text{ kg m}^{-2}$  for savanna grasslands for mid/late dry season burns.

Based on these studies, we roughly estimate that the fuel biomass consumption is  $F = 0.35 \text{ kg}_{\text{dry matter}} \text{ m}^{-2}$ .

## S5 Estimating

In general, the abundance of prescribed burning depends on the accumulation of biomass: the higher the accumulation, the shorter the fire interval is. As a consequence, the fire interval depends on rainfall and grazing pressure, thus showing pronounced regional variability.

For phryganic rangelands in Greece, it is recommended to set fire every 3 to 4 years, which allows to have a good herbage production and at the same time to suppress undesirable dwarf shrub. In South Africa, Oluwole et al. (2008) found that the recovery period should be 3 years for optimum productivity in the absence of grazing. In line with this, the Burning Guidelines of South Africa do not recommend to burn pasture every year (what some farmers do), but every 2-5 years in mesic and coastal grasslands and only when it is needed in dry highveld grasslands. Smith et al. (2013) found that grass richness, evenness, and diversity was high for sites with high rainfall when frequent burning was applied in the dry season (1- to 3-year return intervals), whereas Little et al. (2015) conclude that annual burning combined with intensive grazing has a detrimental effect on plant species diversity and structure. In Australia, single fires caused a short-term reduction of yield and cover of pastures in the following year, but fast recovery occurred for most burning regimes. However, perennial grasses were reduced on the costs of annual grasses, which is why burning every 5-6 and 4-6 years for arid short grass and ribbon blue grass, respectively, are recommended. This is in agreement with the findings of Norman (1963) and Norman (1969) for native pasture on Tippera clay loam in the Katherine region. For North America, the recommended fire return interval of prescribed patch burning is 3 years in areas with rainfall above  $\approx 760 \text{ mm}$  per year and 4 years in drier regions.

One could argue that farmers in the past did not necessarily follow these present-day guidelines. However, traditional knowledge of prescribed burning has been lost in many European areas. Guidelines thus partially re-establish knowledge that our ancestors had. On the one hand, if burning every year reduces the productivity of many grasslands, we think that it is unlikely that ancient farmers conducted burning so often. On the other hand, a too long period without burning is also unlikely since this e.g. allows the growth of unwanted species and can have adverse effects on the ecosystem. According to the summarised literature, burning every  $\approx 3$  years seems to be a reasonable intermediate estimate. Therefore, we assumed that 30% of the pasture area is burnt per year for the intermediate scenario. For the low and the high emission scenarios, we changed this fraction by a factor of 2 and thus arrive at 15% and 60%, respectively.

## S5 Seasonal impact of anthropogenic land cover change

**Tab. S8:** Absolute values of the reference simulation no\_human (“no”) and the simulation LCC\_HYDE (“H”) for each season (JJA=summer, SON=autumn, DJF=winter, and MAM=spring) and the whole year: cloud droplet number concentration burden, liquid water path, cloud cover, cloud radiative effect, ~~precipitation~~, wind velocity at 10 m altitude, surface albedo over land, evaporative fraction, ~~and~~ turbulent flux, ~~land surface temperature~~, and ~~2 m temperature~~. The values are averaged from 10° W to 50° E ~~and from~~, 20° N to 60° N. ~~Changes Relative~~ changes are shown in brackets (~~relative except for temperature~~) and the stars indicate significant changes (5% significance level;  $N = 20$ ).

Var.	Unit	no JJA	H JJA	no SON	H SON	no DJF	H DJF	no MAM	H MAM	no year	H year
CDNC	$10^9 \text{ m}^{-2}$	31.46	30.54 (-2.9%)	27.12	25.66 (-5.4%)	12.85	13.19 (2.6%)	22.49	21.92 (-2.5%)	23.52	22.87 (-2.8%)
LWP	$\text{g m}^{-2}$	54.96	54.89 (-0.1%)	59.03	56.36 (-4.5%)	39.22	39.10 (-0.3%)	41.49	40.13 (-3.3%)	48.69	47.64 (-2.2%)
CC	-	0.32	0.31 (-3.1%)	0.50	0.51 (2.1%)	0.58	0.58 (1.2%)	0.54	0.54 (-0.7%)	0.48	0.48 (0.2%)
CRE	$\text{W m}^{-2}$	-22.44	-22.58 (0.6%)	-4.23	-2.91 (-31.1%)*	4.57	4.60 (0.5%)	-14.43	-14.12 (-2.1%)	-9.21	-8.83 (-4.1%)
Wind <sub>10</sub>	$\text{m s}^{-1}$	4.02	4.05 (0.8%)	4.09	4.14 (1.1%)	4.63	4.66 (0.6%)	4.33	4.37 (1.1%)	4.27	4.30 (0.9%)*
Albedo	-	0.26	0.26 (0.2%)*	0.25	0.25 (-0.0%)	0.26	0.26 (-0.3%)	0.25	0.25 (-0.2%)	0.26	0.26 (-0.1%)
Evap_frac	-	0.36	0.36 (-1.2%)	0.32	0.33 (0.8%)	0.32	0.33 (3.2%)*	0.43	0.43 (0.7%)	0.36	0.36 (0.8%)
F <sub>turb</sub>	$\text{W m}^{-2}$	90.23	89.66 (-0.6%)	64.62	64.80 (0.3%)	52.26	52.73 (0.9%)	79.54	79.98 (0.5%)	71.78	71.91 (0.2%)

**Tab. S9:** The same as Table S8 but for the simulations no\_human and LCC\_KK (“KK”).

Var.	Unit	no JJA	KK JJA	no SON	KK SON	no DJF	KK DJF	no MAM	KK MAM	no year	KK year
CDNC	$10^9 \text{ m}^{-2}$	31.46	29.84 (-5.2%)	27.12	25.51 (-5.9%)*	12.85	11.73 (-8.7%)*	22.49	20.36 (-9.5%)*	23.52	21.90 (-6.9%)*
LWP	$\text{g m}^{-2}$	54.96	53.48 (-2.7%)	59.03	55.95 (-5.2%)*	39.22	36.07 (-8.1%)*	41.49	37.96 (-8.5%)*	48.69	45.89 (-5.8%)*
CC	-	0.32	0.31 (-1.7%)	0.50	0.50 (1.3%)	0.58	0.58 (0.1%)	0.54	0.53 (-1.5%)	0.48	0.48 (-0.3%)
CRE	$\text{W m}^{-2}$	-22.44	-22.20 (-1.1%)	-4.23	-3.13 (-25.8%)*	4.57	4.59 (0.4%)	-14.43	-13.28 (-8.0%)*	-9.21	-8.58 (-6.8%)*
Wind <sub>10</sub>	$\text{m s}^{-1}$	4.02	4.18 (3.9%)*	4.09	4.22 (3.2%)*	4.63	4.74 (2.4%)*	4.33	4.49 (3.7%)*	4.27	4.41 (3.3%)*
Albedo	-	0.26	0.26 (0.8%)*	0.25	0.25 (0.2%)	0.26	0.26 (0.0%)	0.25	0.25 (-0.5%)*	0.26	0.26 (0.1%)
Evap_frac	-	0.36	0.36 (1.1%)	0.32	0.33 (1.3%)	0.32	0.33 (3.9%)*	0.43	0.43 (0.6%)	0.36	0.36 (1.6%)*
F <sub>turb</sub>	$\text{W m}^{-2}$	90.23	90.20 (-0.0%)	64.62	64.13 (-0.8%)	52.26	52.30 (0.1%)	79.54	80.87 (1.7%)	71.78	71.99 (0.3%)

## **S6 Comparison of OC and SO<sub>2</sub> emissions from anthropogenic aerosol sources and natural fire emissions in around AD 100 and AD 1850**

To put our calculated anthropogenic aerosol emissions for AD 100 into some temporal context, we compare them to the anthropogenic aerosol emissions in AD 1850 based on the ACCMIP (Atmospheric Chemistry & Climate Model Intercomparison Project) inventory. The emissions are in both cases averaged over our study domain and the year. ACCMIP includes the sectors energy production and distribution, industry, land transport, maritime transport, residential and commercial combustion, agricultural waste burning on fields, and waste. Not all of these sectors produced aerosol emissions in AD 100; our emissions thus only include fuel consumption (both due to industrial as well as residential combustion), agricultural waste burning on fields, and pasture burning. The emissions from pasture burning might be part of the class “open vegetation fires in savanna and grasslands” in ACCMIP and are thus not considered in this comparison; we expect that the ACCMIP emissions in AD 1850 could be somewhat higher if pasture burning were included.

The BC emissions from the high scenario in AD 100 are nearly as high as the emissions in AD 1850, whereas the emissions from the low and the intermediate scenarios are considerably lower (Fig. ??a). For OC, the emissions for the low and the intermediate scenarios are lower than the emissions in AD 1850, whereas the high scenario results in approximately twice as high aerosol emissions as in AD 1850 (Fig. ??b). The large OC emissions in AD 100 could be due to differences in emission factors: uncertainties in emission factors from biomass burning are large, and the composition of fuels was different in AD 1850 than in AD 100 (e.g. more coal in AD 1850). For SO<sub>2</sub>, the emissions in AD 100 are for all scenarios clearly lower than those in AD 1850 (Fig. ??a), which might again be related to the larger contribution of fossil fuels in AD 1850.

Comparing the aerosol emissions per capita gives similar results (Fig. ??): relative to AD 1850, emissions are highest in AD 100 for OC, followed by BC and then SO<sub>2</sub>. For OC, the per capita emissions are higher in AD 100 than in AD 1850 for the high and the intermediate scenarios. For BC, the per capita emissions are only higher for the high emission scenario, while the per capita emissions for SO<sub>2</sub> are lower than in AD 1850 for all scenarios.

The annual mean anthropogenic aerosol emissions in AD 100 averaged over the study domain (10° W to 50° E, 20° N to 60° N) for the low emission scenario (low), the intermediate emission scenario (int), and the high emission scenario (high). Also shown are the anthropogenic aerosol emissions from the ACCMIP inventory for the year AD 1850 averaged over the same region. The emissions for (a) BC, (b) OC, and (c) SO<sub>2</sub> are pictured.

The same as Figure ?? but showing per capita emissions:

## **S7 Comparison between natural fire emissions and anthropogenic emissions in AD 100**

In the following, we compare the anthropogenic aerosol emissions to the natural fire emissions in AD 100; if the anthropogenic aerosol emissions are very low compared to the natural fire emissions, we would not expect any influence of the anthropogenic emissions. On the other hand, if the anthropogenic aerosol emissions are on the same order of magnitude as the natural fire emissions or higher, then they could have an impact on radiation and clouds, depending on the amount of natural aerosols from other sources (i.e. dust, sea salt, organic aerosol emissions from biogenic sources, and sulfate from volcanic, oceanic, and terrestrial sources).

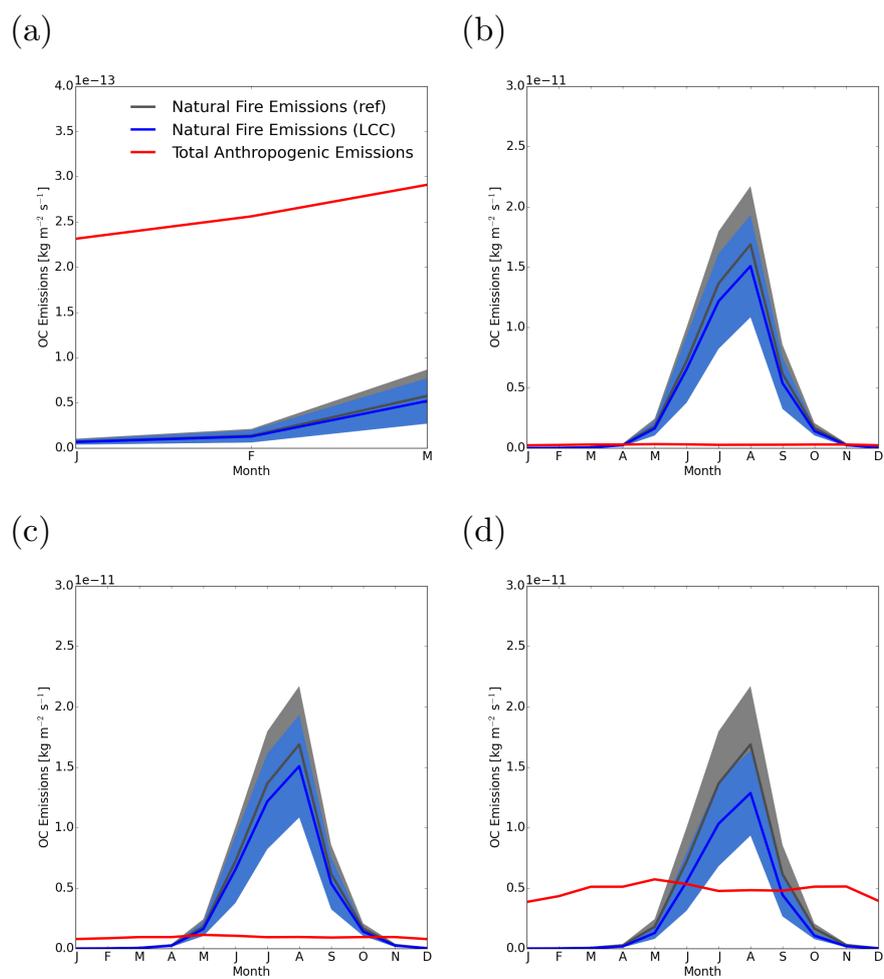
Next to the anthropogenic aerosol emissions, Figs. ??, S2, S3 show also the natural fire emissions used in the simulations without anthropogenic aerosols (no\_human, LCC\_HYDE, LCC\_KK; called “ref” in the figure) and the natural fire emissions for the respective scenarios where the natural fire emissions are set to 0 in areas where crop or pasture grow (LCC\_HYDE\_low, LCC\_HYDE\_int, LCC\_KK\_high; called “LCC” in the figure). By comparing “ref” to the sum of “LCC” and the anthropogenic emissions, the potential impact of the anthropogenic emissions can be estimated.

The anthropogenic emissions have a much less pronounced annual cycle than the natural emissions. Averaged over the whole year, the anthropogenic emissions of the low (Fig. ??b) and the intermediate (Fig. ??e) scenarios are very small compared to natural fire emissions (ref), while the anthropogenic emissions of the high scenario are comparable. However, the comparison strongly depends on the season. For the low emission scenario, the total aerosol emissions from natural fires (“LCC”) plus anthropogenic activities are clearly smaller in summer than the natural fire emissions (“ref”), i.e. humans overall reduce aerosol emissions. In contrast, the anthropogenic emissions are clearly higher than the natural fire emissions in winter, as illustrated in Fig. ??a, which shows the months January to March on a different scale (two orders of magnitude lower). For the intermediate emission scenario, the anthropogenic emissions are larger than the natural fire emissions (“ref”) for approximately half of the year. The anthropogenic emissions for the high emission scenario are higher than the natural fire emissions for most of the year (Fig. ??d).

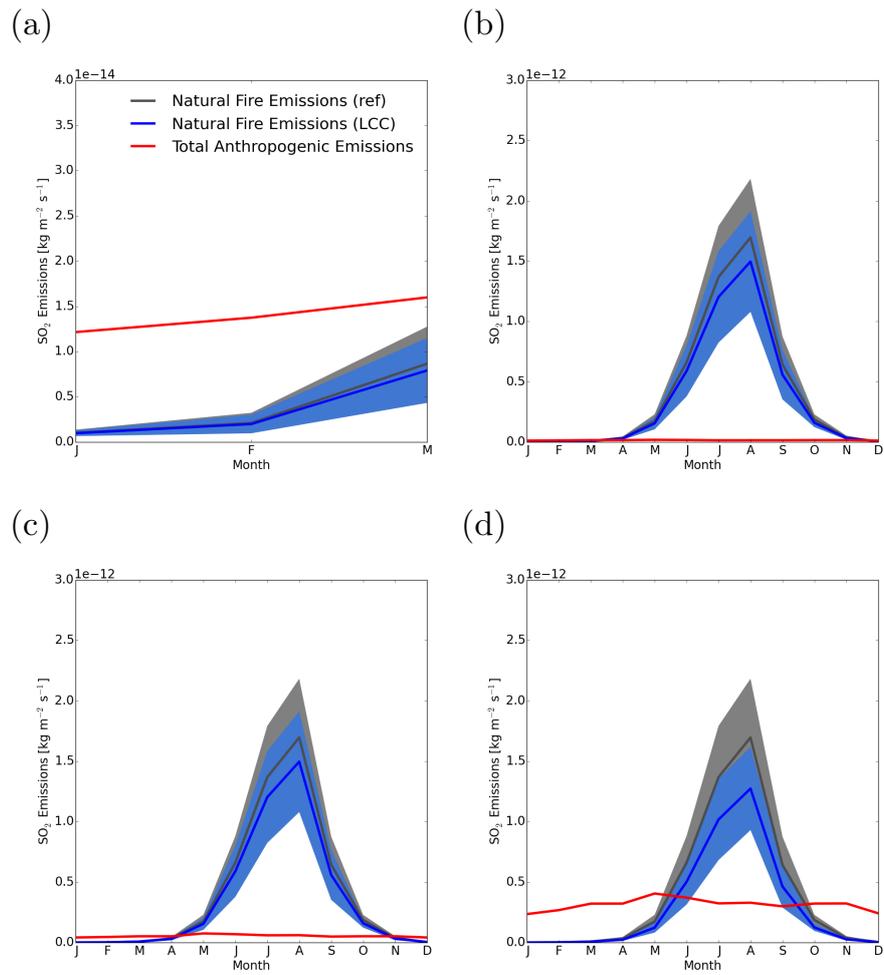
For the majority of scenarios and aerosols, the emissions from pasture burning contribute most to the total anthropogenic emissions (Fig. ??). The emissions from fuel consumption are smaller in most cases but on the same order of magnitude (Fig. ??). Averaged over the whole year, the emissions from crop residue burning are relatively small (Fig. ??); however, in summer, the emissions are in some cases comparable to pasture burning and/or fuel consumption (e.g. Fig. ??b).

Monthly averaged BC emissions in our study domain for (a) the beginning of the year and (b)-(d) the whole year. The shadings show the standard deviation for each month over the 20-year period since our natural fire emissions show interannual variability. The grey line shows the natural fire emissions of the simulations no\_human, LCC\_HYDE, and LCC\_KK, whereas the blue line shows the natural fire emissions of the simulations LCC\_HYDE\_low, LCC\_HYDE\_int, and LCC\_KK\_high, in which anthropogenic land cover change reduces the natural fire emissions. The red line shows the total anthropogenic aerosol emissions. In (a) and (b), the BC emissions from the low scenario are shown; note that the values on the y-axis are larger in (b) than in (a). In (c) and (d), the emissions from the intermediate and the high scenarios are shown, respectively.

Anthropogenic aerosol emissions due to fuel consumption, crop residue burning, and pasture burning in the study domain over the year. (a)-(c): BC; (d)-(f): OC; (g)-(i): SO<sub>2</sub> emissions. Left: low emission scenario; middle: intermediate emission scenario; right: high emission scenario. Note the different scales on the y-axis.



**Fig. S2:** The same as Fig. ?? 4 in the manuscript but for OC instead of BC.



**Fig. S3:** The same as Fig. ??-4 in the manuscript but for  $\text{SO}_2$  instead of BC.

## S7 Seasonal impact of anthropogenic aerosol emissions

**Tab. S10:** Absolute values of the simulation LCC\_HYDE and the simulation LCC\_HYDE\_low (“low”) for each season (JJA=summer, SON=autumn, DJF=winter, and MAM=spring) and the whole year for the following variables: black carbon burden, organic matter burden, SO<sub>4</sub> burden, cloud droplet number concentration burden, liquid water path, cloud cover, ~~precipitation~~, aerosol radiative effect, ~~and~~ cloud radiative effect, ~~land surface temperature~~, and ~~2-m-temperature~~. The values are averaged from 10° W to 50° E and from 20° N to 60° N. ~~Changes-Relative changes~~ are shown in brackets (~~relative except for temperature~~) and the stars indicate significant changes (5% significance level;  $N = 20$ ).

Var.	Unit	H JJA	low JJA	H SON	low SON	H DJF	low DJF	H MAM	low MAM	H year	low year
BC burden	$\mu\text{g m}^{-2}$	538.37	437.39 (-18.8%)*	170.43	141.04 (-17.2%)*	2.72	5.70 (109.7%)*	26.46	26.55 (0.3%)	185.42	153.43 (-17.3%)*
OM burden	$\mu\text{g m}^{-2}$	10151.91	8701.79 (-14.3%)*	3320.51	3051.25 (-8.1%)*	677.58	680.64 (0.5%)	1242.32	1254.44 (1.0%)	3865.01	3436.56 (-11.1%)*
SO <sub>4</sub> burden	$\mu\text{g m}^{-2}$	6331.80	6055.52 (-4.4%)*	4569.16	4596.97 (0.6%)	2600.15	2691.03 (3.5%)	3733.66	3680.67 (-1.4%)	4316.42	4262.86 (-1.2%)
CDNC	$10^9 \text{ m}^{-2}$	30.54	32.02 (4.8%)*	25.66	30.26 (17.9%)*	13.19	17.62 (33.6%)*	21.92	24.45 (11.5%)*	22.87	26.12 (14.2%)*
LWP	$\text{g m}^{-2}$	54.89	56.06 (2.1%)*	56.36	67.22 (19.3%)*	39.10	56.95 (45.7%)*	40.13	46.66 (16.3%)*	47.64	56.69 (19.0%)*
CC	-	0.31	0.32 (4.0%)*	0.51	0.51 (1.5%)	0.58	0.60 (3.1%)*	0.54	0.55 (2.5%)*	0.48	0.50 (2.7%)*
ARE	$\text{W m}^{-2}$	-0.81	-0.76 (-5.9%)*	-0.94	-0.95 (1.5%)	-0.51	-0.47 (-6.8%)*	-0.40	-0.22 (-43.2%)*	-0.66	-0.60 (-9.0%)*
CRE	$\text{W m}^{-2}$	-22.58	-22.78 (0.9%)*	-2.91	-4.96 (70.3%)*	4.60	1.45 (-68.5%)*	-14.12	-17.20 (21.8%)*	-8.83	-10.95 (23.9%)*

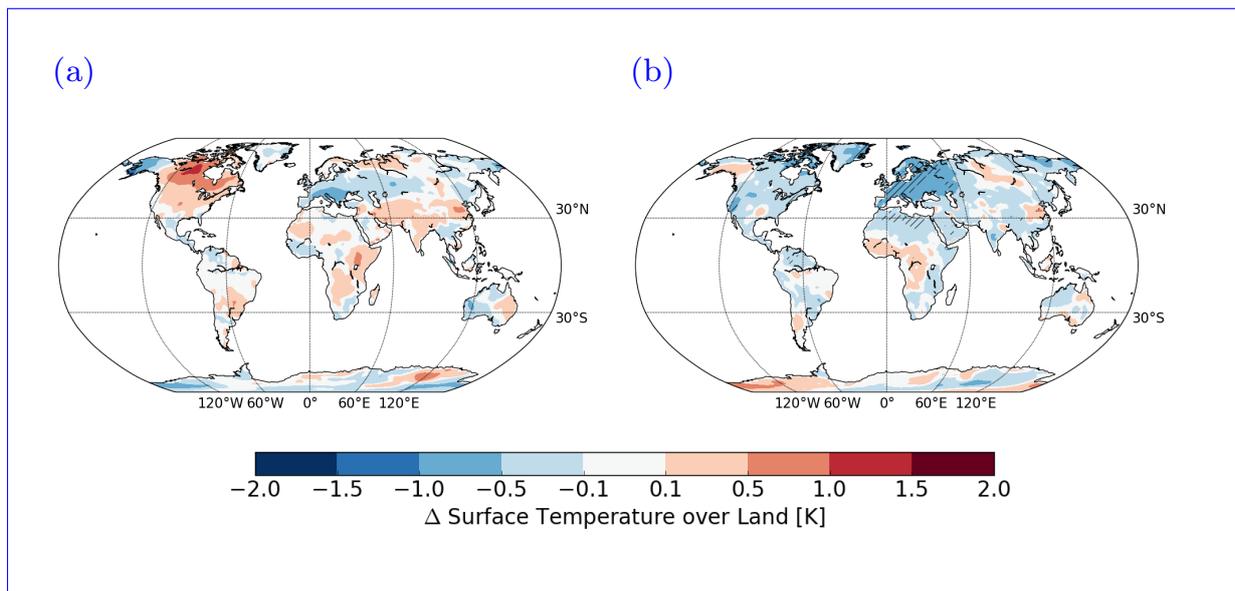
**Tab. S11:** The same as Table S10 but showing results for the simulations LCC\_HYDE\_int (“int”) and LCC\_HYDE.

Var.	Unit	H JJA	int JJA	H SON	int SON	H DJF	int DJF	H MAM	int MAM	H year	int year
BC burden	$\mu\text{g m}^{-2}$	538.37	454.27 (-15.6%)*	170.43	153.68 (-9.8%)*	2.72	14.93 (448.8%)*	26.46	44.12 (66.7%)*	185.42	167.54 (-9.6%)*
OM burden	$\mu\text{g m}^{-2}$	10151.91	8781.79 (-13.5%)*	3320.51	3180.92 (-4.2%)*	677.58	850.66 (25.5%)*	1242.32	1516.03 (22.0%)*	3865.01	3596.95 (-6.9%)*
SO <sub>4</sub> burden	$\mu\text{g m}^{-2}$	6331.80	6067.79 (-4.2%)*	4569.16	4613.68 (1.0%)	2600.15	2682.92 (3.2%)*	3733.66	3682.05 (-1.4%)	4316.42	4268.40 (-1.1%)
CDNC	$10^9 \text{ m}^{-2}$	30.54	34.60 (13.3%)*	25.66	36.36 (41.7%)*	13.19	29.18 (121.2%)*	21.92	30.86 (40.8%)*	22.87	32.76 (43.2%)*
LWP	$\text{g m}^{-2}$	54.89	58.96 (7.4%)*	56.36	76.20 (35.2%)*	39.10	77.44 (98.1%)*	40.13	55.60 (38.5%)*	47.64	66.97 (40.6%)*
CC	-	0.31	0.32 (3.8%)*	0.51	0.53 (3.9%)*	0.58	0.62 (6.1%)*	0.54	0.55 (2.7%)*	0.48	0.50 (4.2%)*
ARE	$\text{W m}^{-2}$	-0.81	-0.79 (-2.8%)*	-0.94	-0.92 (-2.1%)*	-0.51	-0.43 (-16.3%)*	-0.40	-0.05 (-88.6%)*	-0.66	-0.54 (-18.0%)*
CRE	$\text{W m}^{-2}$	-22.58	-24.12 (6.9%)*	-2.91	-5.96 (104.7%)*	4.60	-0.86 (-118.7%)*	-14.12	-20.77 (47.1%)*	-8.83	-13.01 (47.2%)*

**Tab. S12:** The same as Table S10 but showing results for LCC\_KK and LCC\_KK\_high (“high”).

Var.	Unit	KK JJA	high JJA	KK SON	high SON	KK DJF	high DJF	KK MAM	high MAM	KK year	high year
BC burden	$\mu\text{g m}^{-2}$	516.73	520.93 (0.8%)	169.10	233.65 (38.2%)*	2.67	70.53 (2542.2%)*	26.37	131.05 (397.0%)*	179.61	239.89 (33.6%)*
OM burden	$\mu\text{g m}^{-2}$	9466.62	9534.91 (0.7%)	3210.24	4332.61 (35.0%)*	623.35	1600.17 (156.7%)*	1145.62	2634.95 (130.0%)*	3627.28	4540.62 (25.2%)*
SO <sub>4</sub> burden	$\mu\text{g m}^{-2}$	6280.78	6065.80 (-3.4%)	4646.45	4765.02 (2.6%)	2566.28	2820.00 (9.9%)*	3699.46	3729.81 (0.8%)	4305.81	4351.47 (1.1%)
CDNC	$10^9 \text{ m}^{-2}$	29.84	42.75 (43.3%)*	25.51	62.60 (145.4%)*	11.73	67.39 (474.5%)*	20.36	60.85 (198.8%)*	21.90	58.34 (166.4%)*
LWP	$\text{g m}^{-2}$	53.48	63.23 (18.2%)*	55.95	91.92 (64.3%)*	36.07	106.09 (194.2%)*	37.96	77.16 (103.3%)*	45.89	84.47 (84.1%)*
CC	-	0.31	0.33 (6.6%)*	0.50	0.54 (7.6%)*	0.58	0.64 (10.7%)*	0.53	0.58 (8.8%)*	0.48	0.52 (8.7%)*
ARE	$\text{W m}^{-2}$	-0.68	-0.81 (19.9%)	-1.00	-0.97 (-2.9%)	-0.51	-0.40 (-21.5%)*	-0.37	0.10 (-127.3%)*	-0.64	-0.52 (-18.7%)*
CRE	$\text{W m}^{-2}$	-22.20	-25.29 (13.9%)*	-3.13	-7.21 (130.0%)*	4.59	-4.33 (-194.3%)*	-13.28	-27.69 (108.5%)*	-8.58	-16.21 (88.9%)*

## S8 Fixed SST versus MLO



*Fig. S4:* The impact of anthropogenic land cover and aerosol emissions on land surface temperature for the intermediate emission scenario (difference between LCC HYDE\_int and no human). Shown are changes simulated with fixed SST (a) and changes simulated with an MLO (b).

## S9 Comparison between CBALONE-SPITFIRE emissions and emissions by van Marle et al. (2017)

Here, we compare the CBALONE-SPITFIRE emissions ~~for~~ around AD 1835 with the emissions of van Marle et al. (2017) ~~for~~ around AD 1835, which could show biases in the fire model. The reconstructed emissions by van Marle et al. (2017) are used as input for simulations in the context of the Climate Model Intercomparison Project phase 6; we therefore call them “CMIP6 emissions” in the following. The CMIP6 emissions were calculated by merging satellite data with proxies for biomass burning (e.g. charcoal records and visibility observations) next to using the average of six fire models. The emissions simulated by CBALONE-SPITFIRE are not totally independent of the CMIP6 emissions because 3 of the 6 models considered by van Marle et al. (2017) include SPITFIRE (one of them was JSBACH-SPITFIRE).

The global averages of the fire emissions are relatively similar between CBALONE-SPITFIRE and CMIP6 ~~for~~ around AD 1835 (Table S13). The values are somewhat larger for CBALONE-SPITFIRE, namely a factor of 1.2 for  $\text{SO}_2$  and a factor of 1.7 for BC and OC emissions. However, one should keep in mind that the fire emissions by CMIP6 are not the truth and also uncertain. For example, Hamilton et al. (2018) concluded that the CMIP6 pre-industrial fire emissions are likely underestimated.

Although global averages are similar, CBALONE-SPITFIRE simulates pronouncedly higher aerosol emissions compared to CMIP6 ~~for AD 1835~~ in our study domain around AD 1835 (Tables S14, S15, S16). The disagreement is larger in summer and autumn (CBALONE-SPITFIRE 7 to 15 times higher) than in winter and spring (CBALONE-SPITFIRE 2 ~~and to~~ 4 times higher). Furthermore, the fire emissions in winter are more located to the South in CBALONE-SPITFIRE; it simulates basically no fires in Eastern Europe in contrast to CMIP6, but higher

emissions in North Africa and the Middle East (not shown). Note that the majority ( $\approx 80\%$  in AD 1850) of the CMIP6 fire emissions in the study domain originates from agricultural crop waste burning on field, whereas crop area is excluded from burning in CBALONE-SPITFIRE. The CBALONE-SPITFIRE emissions could differ from the CMIP6 emissions over Europe because the first are only model-based, while the latter also include observational data. Furthermore, the models considered by van Marle et al. (2017) were driven by different forcing data than our CBALONE-SPITFIRE simulations.

Overall, the fire emissions calculated with CBALONE-SPITFIRE are **larger-higher** than the CMIP6 emissions in the study domain **in-around** AD 1835. As a consequence, the natural fire emissions calculated for AD 100 could be overestimated in the study domain.

**Tab. S13:** Global mean fire emissions (in  $10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ ) around AD 1835 used for CMIP6 and those calculated with CBALONE-SPITFIRE (“CBAL-SPIT”). The data covers 20 years around AD 1835 (AD 1823-1842).

	CMIP6	CBAL-SPIT
BC	10.0	17.0
OC	84.6	141
SO <sub>2</sub>	12.5	14.5

**Tab. S14:** Mean BC fire emissions (in  $10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ ) averaged from  $10^\circ \text{ W}$  to  $50^\circ \text{ E}$  and from  $20^\circ \text{ N}$  to  $60^\circ \text{ N}$  used for CMIP6 and those calculated with CBALONE-SPITFIRE (“CBAL-SPIT”). The data covers 20 years around AD 1835 (AD 1823-1842).

	CMIP6 AD 1835	CBAL-SPIT AD 1835	CBAL-SPIT AD 100
Autumn	6.65	62.4	33.9
Winter	0.470	1.95	0.339
Spring	6.29	16.7	9.20
Summer	14.8	135	142

**Tab. S15:** The same as Table S14 but for OC and in  $10^{-13} \text{ kg m}^{-2} \text{ s}^{-1}$ .

	CMIP6 AD 1835	CBAL-SPIT AD 1835	CBAL-SPIT AD 100
Autumn	2.80	43.0	26.9
Winter	0.295	1.07	0.178
Spring	3.39	11.1	7.14
Summer	6.77	101	126

**Tab. S16:** The same as Table S14 but for SO<sub>2</sub> (in  $10^{-14} \text{ kg m}^{-2} \text{ s}^{-1}$ ).

	CMIP6 AD 1835	CBAL-SPIT AD 1835	CBAL-SPIT AD 100
Autumn	4.54	40.2	28.8
Winter	0.435	1.57	0.250
Spring	5.06	10.2	7.27
Summer	10.8	78.7	125

Comparing the two CBALONE-SPITFIRE simulations (AD 1835 versus AD 100; Tables S14, S15, S16) reveals that emissions **in-around** AD 1835 were somewhat lower in summer compared to AD 100. However, for the other seasons, the emissions in AD 1835 are higher, especially in winter (a factor of 6). The differences can partly be explained by the differences in the population density; for AD 100, only natural fires were calculated and the population density

was thus set to 0 (Sect. [2.4](#)). In the model, population shortens the fire duration but at the same time increases the number of ignition events. While the first effect dominates in summer, the second one dominates in the other seasons. Furthermore, differences in anthropogenic land cover (none assumed for AD 100), natural vegetation, and climate can also contribute to the differences between AD 1835 and AD 100.

## S10 Emission factors considered for calculation

**Tab. S17:** Compilation of emission factors from burning: wood as fuel (key 1), agricultural residue as fuel (key 2), agricultural residue in the field (key 3), pasture or grass (key 4), charcoal (key 5), and wood for charcoal production (key 6).  $N$  stands for the number of samples, and “a” in the last column indicates for which measurements we estimated the standard deviation (“std”). The values are given in  $\text{g kg}^{-1}$  on a dry matter basis. In this table, we show for simplicity four decimal places for all measurement, but this does not mean that all measurements had this precision.

Burning material	OC		BC		SO <sub>2</sub>		$N$	Reference	Burning/ measurement device	Key
	average	std	average	std	average	std				
UP dung	7.1970	6.7530	0.5657	0.4949			7	Pandey et al. (2017)	traditional cooking-stove, India	2
Bihar dung	10.6500	10.0300	0.9700	0.8403			6	Pandey et al. (2017)	traditional cooking-stove, India	2
Chh rice straw	7.0786	5.1236	1.1157	1.3508			7	Pandey et al. (2017)	traditional cooking-stove, India	2
Chh tur stalk	8.0880	6.1330	1.9580	1.8140			10	Pandey et al. (2017)	traditional cooking-stove, India	2
Punjab wood	2.6000	2.3780	0.6750	0.4387			4	Pandey et al. (2017)	traditional cooking-stove, India	1
Raj wood	5.4041	5.0848	1.1410	1.0134			17	Pandey et al. (2017)	traditional cooking-stove, India	1
UP wood	2.1363	1.1830	0.5500	0.3464			8	Pandey et al. (2017)	traditional cooking-stove, India	1
AP wood	5.0125	3.6248	0.4550	0.2664			4	Pandey et al. (2017)	traditional cooking-stove, India	1
wheat straw	3.4600	2.0500	0.4200	0.2300	0.0400	0.0400	8	Cao et al. (2008)	combustion tower (simulating cooking in traditional stoves)	2
rice straw	2.0100	0.6700	0.4900	0.2100	0.1800	0.3100	14	Cao et al. (2008)	combustion tower (simulating cooking in traditional stoves)	2

corn stover	2.2500	0.7400	0.9500	1.0800	0.0400	0.0400	9	Cao et al. (2008)	combustion tower (simulating cooking in traditional stoves)	2
cotton stalk	1.8300	0.5400	0.8200	0.2000			2	Cao et al. (2008)	combustion tower (simulating cooking in traditional stoves)	2
rice straw	0.9000	0.4500	0.4600	0.2300			7	Turn et al. (1997)	wind tunnel	3a
wheat straw	2.1000	1.0500	0.7900	0.3950			2	Turn et al. (1997)	wind tunnel	3a
barley straw	3.0000	1.5000	1.2000	0.6000			2	Turn et al. (1997)	wind tunnel	3a
corn stover	1.7000	0.8500	0.6700	0.3350			2	Turn et al. (1997)	wind tunnel	3a
sugar cane	1.5000	0.7500	0.5900	0.2950			3	Turn et al. (1997)	wind tunnel	3a
wheat straw	2.8130	0.1470	0.6760	0.0270			4	Li et al. (2017)	combustion stove to simulate open burning	3
corn straw	2.3930	0.3510	0.7780	0.1520			4	Li et al. (2017)	combustion stove to simulate open burning	3
rice straw	6.8820	0.6890	2.1820	0.2780			4	Li et al. (2017)	combustion stove to simulate open burning	3
cotton residue	7.4150	0.5470	1.1920	0.1710			4	Li et al. (2017)	combustion stove to simulate open burning	3
soybean residue	1.5390	0.2530	0.6140	0.1900			4	Li et al. (2017)	combustion stove to simulate open burning	3
Maize-brick					0.0163	0.0104	3	Zhang et al. (2000)	brick stove with flue	2
Wood-India					0.0024	0.0024	3	Zhang et al. (2000)	metal stove without flue (from India)	1
Wheat-brick					0.0335	0.0174	3	Zhang et al. (2000)	brick stove with flue	2
Brush-brick					0.0056	0.0077	3	Zhang et al. (2000)	brick stove with flue	2
Brush-India					0.0025	0.0044	3	Zhang et al. (2000)	metal stove without flue (from India)	2

fireplace, soft-wood	3.0073	0.3335	0.7742	0.0697			3	McDonald et al. (2000)	heatilator model E36 fireplace	1
fireplace, hard-wood	3.5800	0.7981	0.3975	0.1177			3	McDonald et al. (2000)	heatilator model E36 fireplace	1
wood stove, hard-wood	2.8211	1.7706	0.3563	0.2169			3	McDonald et al. (2000)	noncatalytic Pineridge appliance	1
maritime pine	6.1344	3.0672	1.0650	0.5325			3	Alves et al. (2011)	traditional Portuguese brick open fireplace	1a
eucalypt	8.7264	4.3632	0.4444	0.2222			3	Alves et al. (2011)	traditional Portuguese brick open fireplace	1a
cork oak	6.9412	3.4706	0.3484	0.1742			3	Alves et al. (2011)	traditional Portuguese brick open fireplace	1a
golden wattle	3.8700	1.9350	0.6600	0.3300			3	Alves et al. (2011)	traditional Portuguese brick open fireplace	1a
olive	4.8015	2.4008	0.4158	0.2079			3	Alves et al. (2011)	traditional Portuguese brick open fireplace	1a
Portugese oak	9.1680	4.5840	0.4775	0.2388			3	Alves et al. (2011)	traditional Portuguese brick open fireplace	1a
holm oak	8.4800	4.2400	0.3520	0.1760			3	Alves et al. (2011)	traditional Portuguese brick open fireplace	1a
maritime pine	8.0196	4.0098	0.6357	0.3179			3	Alves et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1a
eucalypt	3.2160	1.6080	0.2479	0.1240			3	Alves et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1a
cork oak	8.0936	4.0468	0.2869	0.1435			3	Alves et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1a
golden wattle	5.2650	2.6325	0.2340	0.1170			3	Alves et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1a

olive	2.8272	1.4136	0.4774	0.2387			3	Alves et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1a
Portugese oak	4.8608	2.4304	0.3822	0.1911			3	Alves et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1a
holm oak	5.4468	2.7234	0.2244	0.1122			3	Alves et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1a
Quercus pyrenaica	9.0000	3.0000	0.5790	0.0890			2	Calvo et al. (2015)	traditional Portuguese brick open fireplace	1
Pinus nigra	10.5000	3.4000	0.7900	0.1900			2	Calvo et al. (2015)	traditional Portuguese brick open fireplace	1
Facus sylvatica	3.9000	0.9200	0.4300	0.2300			2	Calvo et al. (2015)	traditional Portuguese brick open fireplace	1
Quercus pyrenaica	9.1000	2.7000	0.9000	0.2000			6	Calvo et al. (2015)	a cast iron woodstove (Solzaima, model Sahara)	1
Pinus nigra	2.9000	1.1000	0.8790	0.0480			6	Calvo et al. (2015)	a cast iron woodstove (Solzaima, model Sahara)	1
Facus sylvatica	1.6000	0.5000	0.4300	0.1300			6	Calvo et al. (2015)	a cast iron woodstove (Solzaima, model Sahara)	1
tur (woody stalks)	1.6830		0.4080				1	Habib et al. (2008)	U-shaped, single-pot mud stove	1
cotton (woody stalks)	1.3440		0.6720				1	Habib et al. (2008)	U-shaped, single-pot mud stove	1
soyabean (woody stalks)	4.2680		0.7760				1	Habib et al. (2008)	U-shaped, single-pot mud stove	1
mustard (woody stalks)	4.7960		0.6540				1	Habib et al. (2008)	U-shaped, single-pot mud stove	1
cattle dung	2.3220		0.2160				1	Habib et al. (2008)	U-shaped, single-pot mud stove	2

jute (fibrous hol- low stalks)	0.6290		0.3060			1	Habib et al. (2008)	U-shaped, single-pot stove	mud	2
rice straw	4.6500		0.1860			1	Habib et al. (2008)	U-shaped, single-pot stove	mud	2
jamun	1.5800	2.0300	0.4800	0.0850		2	Habib et al. (2008)	U-shaped, single-pot stove	mud	1
neem	1.6500	1.9500	0.6100	0.0300		2	Habib et al. (2008)	U-shaped, single-pot stove	mud	1
mango	1.6100	1.5700	0.6400	0.0640		2	Habib et al. (2008)	U-shaped, single-pot stove	mud	1
Acacia	1.1000	0.5390	0.4500	0.1220		2	Habib et al. (2008)	U-shaped, single-pot stove	mud	1
horsebean	1.2000	0.1700	1.2800	0.1000		2	Shen et al. (2010)	brick wok stove (rural)		2
peanut	1.0800		0.4930			1	Shen et al. (2010)	brick wok stove (rural)		2
soybean residue	1.1700	0.1800	1.3700	0.0400		2	Shen et al. (2010)	brick wok stove (rural)		2
cotton residue	0.3540	0.0410	1.3400	0.5800		2	Shen et al. (2010)	brick wok stove (rural)		2
rice	1.5000	0.1600	0.7490	0.4270		2	Shen et al. (2010)	brick wok stove (rural)		2
wheat	2.2700	1.3000	2.6400	1.0100		2	Shen et al. (2010)	brick wok stove (rural)		2
rape	1.7500	0.6300	2.3400	0.9200		2	Shen et al. (2010)	brick wok stove (rural)		2
sesame	2.3400	1.1200	1.0700	0.0300		2	Shen et al. (2010)	brick wok stove (rural)		2
corn	1.3600	0.2900	1.1100	0.0800		2	Shen et al. (2010)	brick wok stove (rural)		2
wheat stubble	1.3794	0.3012	0.4260	0.1147		3	Jimenez et al. (2007)	test burn chamber to mimic open burn conditions		3
wheat stubble	2.8120	1.6127	0.1660	0.1408		6	Jimenez et al. (2007)	open field		3
Kentucky Blue- grass	5.2248	2.2406	0.7650	0.3361		2	Jimenez et al. (2007)	open field		4
Kentucky Blue- grass	4.4637	1.2962	0.8155	0.4307		4	Jimenez et al. (2007)	test burn chamber to mimic open burn conditions		4

wheat straw	0.2900	0.1200	0.1600	0.0700				Sahai et al. (2007)	field burning	3
wheat straw	2.3800		1.5900				1	Sahai et al. (2007)	earlier lab experiments at NPL	3
wheat straw	1.2300		0.5200				1	Hays et al. (2005)	field open burn simulations	3
rice straw	8.9400		0.1700				1	Hays et al. (2005)	field open burn simulations	3
olive pits	0.9000	0.5700	0.1000	0.0600				AIRUSE LIFE (2016)	from pellet stove	2
maritime pine	2.5400	2.5100	0.6100	0.4300			3	Fernandes et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1
golden wattle	4.0700	2.6500	0.2900	0.1800			3	Fernandes et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1
eucalypt	5.1600	4.0300	0.3700	0.3000			3	Fernandes et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1
cork oak	4.8000	3.3800	0.4200	0.3300			3	Fernandes et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1
olive	4.5500	2.2200	0.4600	0.2400			3	Fernandes et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1
holm oak	3.0300	2.0500	0.2300	0.0900			3	Fernandes et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1
Portugese oak	6.1700	4.6200	0.3200	0.1500			3	Fernandes et al. (2011)	a cast iron woodstove (Solzaima, model Sahara)	1
maritime pine	2.9100	1.3000	0.6200	0.4900			3	Fernandes et al. (2011)	traditional Portuguese brick open fireplace	1
golden wattle	3.5300	3.1300	0.3400	0.2600			3	Fernandes et al. (2011)	traditional Portuguese brick open fireplace	1
eucalypt	5.1100	3.9000	0.3600	0.3600			3	Fernandes et al. (2011)	traditional Portuguese brick open fireplace	1

cork oak	10.0600	5.2400	0.6800	0.4000			3	Fernandes et al. (2011)	traditional Portuguese brick open fireplace	1
olive	9.1000	5.7400	0.3900	0.1600			3	Fernandes et al. (2011)	traditional Portuguese brick open fireplace	1
holm oak	7.2200	4.0300	0.3000	0.1100			3	Fernandes et al. (2011)	traditional Portuguese brick open fireplace	1
Portugese oak	6.0600	3.4000	0.3200	0.2000			3	Fernandes et al. (2011)	traditional Portuguese brick open fireplace	1
Chinese white poplar	0.6600	0.3200	0.8800	0.4900			3	Guofeng et al. (2012)	brick stove (rural)	1
elm	0.7900	0.1500	1.2000	0.3000			3	Guofeng et al. (2012)	brick stove (rural)	1
yellow locust	1.9000	1.5000	0.2100	0.1500			3	Guofeng et al. (2012)	brick stove (rural)	1
maple	0.1100	0.0100	0.0560	0.0040			3	Guofeng et al. (2012)	brick stove (rural)	1
fir	0.9700	0.8800	0.9500	0.2000			3	Guofeng et al. (2012)	brick stove (rural)	1
larch	0.1400	0.1100	0.3500	0.3400			3	Guofeng et al. (2012)	brick stove (rural)	1
water Chinese fir	0.3600	0.1700	0.8500	0.4500			3	Guofeng et al. (2012)	brick stove (rural)	1
cypress	0.8200	0.4500	0.7100	0.3900			3	Guofeng et al. (2012)	brick stove (rural)	1
oak	0.5400	0.6300	0.1300	0.1300			3	Guofeng et al. (2012)	brick stove (rural)	1
chinese pine	0.6000	0.3500	0.9400	0.4000			3	Guofeng et al. (2012)	brick stove (rural)	1

willow	0.2300	0.1000	0.4700	0.3000			3	Guofeng et al. (2012)	brick stove (rural)	1
Paulownia tomentosa	0.3900	0.1200	0.9400	0.5100			3	Guofeng et al. (2012)	brick stove (rural)	1
toon	0.1900	0.1300	0.5200	0.4100			3	Guofeng et al. (2012)	brick stove (rural)	1
white birch	0.6900	0.3200	0.6700	0.6100			3	Guofeng et al. (2012)	brick stove (rural)	1
Lespedeza	0.2100	0.1700	0.4800	0.4000			3	Guofeng et al. (2012)	brick stove (rural)	1
Buxus sinica	1.2000	0.1000	2.5000	1.7000			3	Guofeng et al. (2012)	brick stove (rural)	1
holly	0.7300	0.2800	1.6000	0.4000			3	Guofeng et al. (2012)	brick stove (rural)	1
bamboo	0.1300	0.0900	1.2000	0.5000			3	Guofeng et al. (2012)	brick stove (rural)	1
barley straw					0.0400	0.0000	2	Jenkins et al. (1996); a932-126b_1	wind tunnel	3
corn stover					0.2000	0.0141	2	Jenkins et al. (1996); a932-126b_1	wind tunnel	3
rice straw					0.5910	0.2150	8	Jenkins et al. (1996); a932-126b_1	wind tunnel	3
wheat straw					0.4700	0.2758	2	Jenkins et al. (1996); a932-126b_1	wind tunnel	3

seasoned oak					0.1671		1	DeAngelis et al. (1980)	nonbaffled stove	1
seasoned pine					0.2531		1	DeAngelis et al. (1980)	nonbaffled stove	1
Kentucky Blue-grass	15.0000	5.2000	0.5000	0.1100			4	Holder et al. (2017)	field measurements, Aerostat	4
Kentucky Blue-grass	11.0000	6.1000	0.3200	0.1000			4	Holder et al. (2017)	field measurements, ground	4
Kentucky Blue-grass			0.7600	0.3300			4	Holder et al. (2017)	field measurements, Aerostat, eBC	4
Kentucky Blue-grass			0.9300	0.0730			4	Holder et al. (2017)	field measurements, ground, eBC	4
wheat	9.4000	1.6000	0.5000	0.2000			4	Holder et al. (2017)	field measurements, Aerostat	3
wheat	11.2000	2.5000	0.2000	0.0000			4	Holder et al. (2017)	field measurements, ground	3
wheat			0.6000	0.1000			4	Holder et al. (2017)	field measurements, Aerostat, eBC	3
wheat			0.5000	0.1000			4	Holder et al. (2017)	field measurements, ground, eBC	3
wheat			0.7000	0.0900			8	Zhang et al. (2015)	field measurements, EF based on CO <sub>2</sub>	3
rice			0.5600	0.0400			4	Zhang et al. (2015)	field measurements, EF based on CO <sub>2</sub>	3
rapeseed			2.8900	0.7000			4	Zhang et al. (2015)	field measurements, EF based on CO <sub>2</sub>	3
wheat			0.4300	0.1000			8	Zhang et al. (2015)	field measurements, EF based on CO	3

rice			0.2500	0.1100			4	Zhang et al. (2015)	field measurements, EF based on CO	3
rapeseed			1.0100	0.2700			4	Zhang et al. (2015)	field measurements, EF based on CO	3
charcoal burning	1.7800	2.8000	0.6500	0.3000			8	Keita et al. (2018)	trad. stoves (metal or baked earth), field meas.; per kg charcoal	5
charcoal making	3.9300	1.0100	0.2200	0.1600			8	Keita et al. (2018)	trad. stoves (metal or baked earth), field meas.; per kg wood	6
wood (iroko)	6.5000	1.9800	0.5200	0.3900			4	Keita et al. (2018)	traditional charcoal-making furnaces, field meas.	1
wood(hevea)	15.6100	6.4400	1.4500	0.6100			4	Keita et al. (2018)	traditional charcoal-making furnaces, field meas.	1
charcoal making					0.1860	0.1350	7	Park et al. (2013)	charcoal kiln, field meas.; per kg wood	6
wheat stubble	1.9000	1.0482					6	Dhammapala et al. (2007a)	test burn facility	3
Kentucky Blue-grass stubble	6.9000	2.2346					29	Dhammapala et al. (2007a)	test burn facility	4
wheat stubble			0.3500	0.2382			11	Dhammapala et al. (2007a)	test burn facility	3
Kentucky Blue-grass stubble			0.6300	0.1472			29	Dhammapala et al. (2007a)	test burn facility	4
wheat stubble	2.7360	1.0080					4	Dhammapala et al. (2007b)	field burning	3
Kentucky Blue-grass stubble	6.9330						1	Dhammapala et al. (2007b)	field burning	4

wheat stubble			0.3760	0.2890			6	Dhammapala et al. (2007b)	field burning	3
Kentucky Blue-grass stubble			0.7980	0.3440			2	Dhammapala et al. (2007b)	field burning	4
grass, ground	7.0000		0.6200				1	Strand et al. (2016)	ground	4
grass, aerostat	6.5000		0.5600				1	Strand et al. (2016)	Aerostat	4
grass, ground			1.1000				1	Strand et al. (2016)	ground; continuous eBC	4
grass, aerostat			0.9100				1	Strand et al. (2016)	Aerostat; continuous eBC	4
grass (Brazil)	5.1000	2.2500	0.6500	0.4500	0.4400	0.1800	6	Ferek et al. (1998)	airborne measurements	4
fuel wood, Delhi	0.8500	0.7000	0.3700	0.1800			101	Saud et al. (2012)	dilution sampler	1
fuel wood, Punjab	0.9300	0.9600	0.4400	0.2900			139	Saud et al. (2012)	dilution sampler	1
fuel wood, Haryana	0.7800	0.4100	0.4200	0.0700			92	Saud et al. (2012)	dilution sampler	1
fuel wood, Uttar Pradesh	1.3200	1.0000	0.3100	0.1200			149	Saud et al. (2012)	dilution sampler	1
fuel wood, Uttarakhand	0.9200	0.5400	0.2700	0.0800			181	Saud et al. (2012)	dilution sampler	1
fuel wood, Bihar	1.2800	0.9900	0.3600	0.1900			85	Saud et al. (2012)	dilution sampler	1
fuel wood, West Bengal	0.5500	0.7000	0.2500	0.1300			19	Saud et al. (2012)	dilution sampler	1
crop residue, Delhi	2.1000	1.4400	0.5700	0.3600			20	Saud et al. (2012)	dilution sampler	2
crop residue, Punjab	0.5600	0.2800	0.2500	0.0700			40	Saud et al. (2012)	dilution sampler	2

crop residue, Haryana	0.9400	0.7100	0.4200	0.1300			35	Saud et al. (2012)	dilution sampler	2
crop residue, Uttar Pradesh	2.3400	1.2800	0.3900	0.1900			107	Saud et al. (2012)	dilution sampler	2
crop residue, Bihar	1.8700	1.6600	0.4300	0.5200			105	Saud et al. (2012)	dilution sampler	2
crop residue, West Bengal	0.9700	0.8800	0.1800	0.1100			20	Saud et al. (2012)	dilution sampler	2
dung cake, Delhi	4.5100	1.3400	0.9000	0.3500			95	Saud et al. (2012)	dilution sampler	2
dung cake, Punjab	4.6400	0.7800	0.5900	0.2400			48	Saud et al. (2012)	dilution sampler	2
dung cake, Haryana	3.7800	0.4700	0.5400	0.3400			38	Saud et al. (2012)	dilution sampler	2
dung cake, Uttar Pradesh	4.4100	1.1900	0.4100	0.2100			45	Saud et al. (2012)	dilution sampler	2
dung cake, Bihar	4.1400	1.4800	0.2800	0.2100			68	Saud et al. (2012)	dilution sampler	2
dung cake, West Bengal	1.7400	1.1000	0.2200	0.1300			8	Saud et al. (2012)	dilution sampler	2
rice straw	2.7838	1.1874	0.4789	0.2891			4	Oanh et al. (2011)	field burning	3
wheat straw	2.6000	1.8000	0.4600	0.1900			3	Wei et al. (2014)	improved two-pot stove with chimney, 1-year old	2
rape straw	0.5800	0.1700	0.4700	0.4800			3	Wei et al. (2014)	improved two-pot stove with chimney, 1-year old	2
rice straw	1.1000	0.9000	0.5100	0.3700			3	Wei et al. (2014)	improved two-pot stove with chimney, 1-year old	2
cotton straw	3.1000	4.5000	1.2000	1.4000			3	Wei et al. (2014)	improved two-pot stove with chimney, 1-year old	2
wheat straw	4.1000	1.8000	1.4000	0.9000			3	Wei et al. (2014)	improved two-pot stove with chimney, 15-year old	2

rape straw	4.9000	6.5000	2.7000	2.4000			3	Wei et al. (2014)	improved two-pot stove with chimney, 15-year old	2
rice straw	1.5000	0.6000	0.6400	0.1900			3	Wei et al. (2014)	improved two-pot stove with chimney, 15-year old	2
cotton straw	2.5000	1.8000	0.9600	0.9000			3	Wei et al. (2014)	improved two-pot stove with chimney, 15-year old	2
wheat straw	4.0659	0.3297	0.3187	0.0220			3	Tian et al. (2017)	traditional stove	2
maize straw	7.0330	1.9780	0.5275	0.0769			3	Tian et al. (2017)	traditional stove	2
rice straw	8.4615	1.4286	0.5714	0.0659			3	Tian et al. (2017)	traditional stove	2
charcoal burning					0.5000	0.3000	1	Gadi et al. (2011)	U-shaped chimney	5

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