

Fire and biofuel contributions to annual mean aerosol mass concentrations in the United States

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Abstract

We estimate the contributions from biomass burning (summer wildfires, other fires, residential biofuel, and industrial biofuel) to seasonal and annual aerosol concentrations in the United States. Our approach is to use total carbonaceous (TC) and non-soil potassium (ns-K) aerosol mass concentrations for 2001–2004 from the nationwide IMPROVE network of surface sites, together with satellite fire data. We find that summer wildfires largely drive the observed interannual variability of TC aerosol concentrations in the United States. TC/ns-K mass enhancement ratios from fires range from 10 for grassland and shrub fires in the south to 130 for forest fires in the north. The resulting summer wildfire contributions to annual TC aerosol concentrations for 2001–2004 are $0.26 \mu\text{g C m}^{-3}$ in the west and $0.14 \mu\text{g C m}^{-3}$ in the east; Canadian fires are a major contributor in the east. Non-summer wildfires and prescribed burns contribute on an annual mean basis 0.27 and $0.31 \mu\text{g C m}^{-3}$ in the west and the east, highest in the southeast because of prescribed burning. Residential biofuel is a large contributor in the northeast with annual mean concentration of up to $2.2 \mu\text{g C m}^{-3}$ in Maine. Industrial biofuel (mainly paper and pulp mills) contributes up to $0.3 \mu\text{g C m}^{-3}$ in the southeast. Total annual mean fine aerosol concentrations from biomass burning average 1.2 and $1.6 \mu\text{g m}^{-3}$ in the west and east, respectively, contributing about 50% of observed annual mean TC concentrations in both regions and accounting for 30% (west) and 20% (east) of total observed fine aerosol concentrations. Our analysis supports bottom-up source estimates for the contiguous United States of $0.7\text{--}0.9 \text{ Tg C yr}^{-1}$ from open fires (climatological) and 0.4 Tg C yr^{-1} from biofuel use. Biomass burning is thus an important contributor to US air quality degradation, which is likely to grow in the future.

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1. Introduction

Fires are an important but poorly quantified factor for air quality degradation in the United States. Carbonaceous aerosol (smoke) emissions are of most concern. Episodic effects are well established, for example, from the wildfires that occurred in California and Oregon in the summer of 2002 (McMeeking et al., 2005; Malm et al., 2005; Carrico

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et al., 2005) or in Southern California in October 2003 (Westerling et al., 2004; Phuleria et al., 2005). There is also significant transboundary pollution influence from fires in Canada (Wotawa and Trainer, 2000; McKeen et al., 2002; Colarco et al., 2004; DeBell et al., 2004) and in Mexico (Peppler et al., 2000; Kreidenweis et al., 2001; Wang et al., 2006). However, little attention has been paid to the more diffuse, nationwide effects of fires on seasonal and annual aerosol concentrations of relevance for air quality and visibility standards. Fire emissions are often not included in regional air quality models (e.g., Byun and Ching, 1999). Effects from biofuel use, both residential and industrial, have also received little attention. We address these issues here using aerosol composition measurements from the IMPROVE nationwide network of remote surface sites for 2001–2004 (Malm et al., 1994).

Fires in the United States include wildfires as well as prescribed agricultural and land management burns. The former are the largest contributor to the annual total area burned and occur largely in the west and Alaska (US EPA, 2003a). By contrast, over 70% of all prescribed burning is in the southeast (Hardy et al., 2001). The largest wildfires occur in conifer forests, but grassland and shrub fires are also important. Wildfires are naturally initiated by lightning but human negligence and arson are also frequent causes (Hardy et al., 2001). The US wildfire season generally extends from May to October and peaks in June–August when temperatures are highest (Westerling et al., 2003). Fire intensity varies considerably from year to year and is strongly associated with hot and dry weather conditions (Westerling and Swetnam, 2003). Westerling et al. (2006) present evidence for an increase in large fires since the 1970s in the western United States, associated with increasing spring and summer temperatures. An increase in wildfires is expected in the future because of accumulated stocks and climate change (Brown et al., 2004; Soja et al., 2006).

Burning of biofuels such as wood, charcoal, and agricultural residues provides an important source of energy for residential and industrial sectors in developing countries (Yevich and Logan, 2003). In the United States, ~80% of biofuels are consumed by agro-industrial factories such as paper and pulp mills and the rest is used for residential heating (Energy Information Administration, 2006).

In previous work (Park et al., 2003), we estimated the contributions from different source types to

annual mean carbonaceous aerosol concentrations in the United States. We used the GEOS-Chem chemical transport model (CTM) to simulate observations in 1998 at the IMPROVE sites, and adjusted emissions in the model to match the spatial and seasonal patterns in the observations. We then generalized our results by using the climatological fire inventory of Duncan et al. (2003). We found that fires contributed 0.48 and 0.32 $\mu\text{g C m}^{-3}$ to mean carbonaceous aerosol concentrations for the western and eastern United States, respectively. We also found that the biofuel contribution to annual mean organic carbon aerosol concentrations is larger than that of the fossil fuel on a national scale (0.47 $\mu\text{g C m}^{-3}$ vs. 0.29 $\mu\text{g C m}^{-3}$).

These fire and biofuel contributions to aerosol concentrations have implications not only for health-based air quality standards but also for visibility. The Regional Haze Rule of the US Environmental Protection Agency (EPA) aims to achieve linear improvement in visibility in wilderness areas at a rate designed to meet an aspirational endpoint of natural visibility conditions by 2064 (US EPA, 2003b). Phase 1 of the Rule mandates states to achieve this linear improvement over the 2004–2018 period. As discussed by Park et al. (2004, 2006), the definition of natural visibility has immediate impact on the 2004–2018 emission reduction schedule because visibility degradation is a logarithmic function of aerosol concentrations. Better quantification of fire contributions is thus crucial in the application of the Rule (Park et al., 2006).

In this paper, we use an observationally based analysis of the IMPROVE data for 2001–2004 to quantify the contributions from fires and biofuels to carbonaceous aerosol concentrations in the United States, and the associated geographical and inter-annual variability. Our analysis involves correlations of carbonaceous aerosol concentrations with non-soil potassium together with fire information. We discuss the implications of our findings for the health-based national ambient air quality standard (NAAQS) as well as for the Regional Haze Rule.

2. Methods

The IMPROVE monitoring program was initiated in 1987 in national parks and other protected environments to identify the contributions of different aerosol components to visibility degradation. As of 2001, it included >100 sites nationwide at locations that have little impact from nearby sources. The data

consist of 24-h speciated fine aerosol ($<2.5\mu\text{m}$ in diameter) mass concentrations measured every third day including organic carbon (OC), elemental carbon (EC), potassium (K), iron (Fe), and other components (Malm et al., 1994). K and Fe are measured by particle-induced X-ray emission and X-ray fluorescence, respectively. OC and EC are determined using the Thermal Optical Reflectance method, in which some of the refractory OC may be measured as EC (Chow et al., 1993). We refer to the sum of OC and EC as the total carbonaceous (TC) aerosol concentration, which is a more robust measurement (Chow et al., 2001; Venkatachari et al., 2006). Organic carbon accounts for $\sim 90\%$ of TC in fire plumes (Andreae and Merlet, 2001; McMeeking et al., 2005). Uncertainties with IMPROVE measurements of TC, K, and Fe are typically $<10\%$ (<http://vista.cira.colostate.edu/>).

Potassium (K) is a crustal element present in soil dust and in all living organisms (Mason, 1982). It is emitted to the atmosphere by biomass burning but not by fossil fuel combustion (Andreae, 1983). We use the Fe fine aerosol data at the IMPROVE sites as a surrogate for soil K with an estimated K/Fe mass ratio of 0.6 ± 0.2 (Cahill et al., 1986; Malm et al., 1994). After removing soil K from the observed K concentration, we refer to the remaining K as non-soil potassium (ns-K) to trace the biomass burning influence (Ma et al., 2003; Kreidenweis et al., 2001; Wang et al., 2006). “Biomass burning” is taken here to include both open fires and biofuel combustion.

Fig. 1 shows monthly mean ns-K concentrations at illustrative western (Yellowstone, Wyoming) and eastern (Acadia, Maine) IMPROVE sites for 2001–2004. Values at Yellowstone are highest in summer and very low in winter, reflecting the

influence of wildfires in summer and negligible burning in winter. Values at Acadia are significant throughout the year and are in general high in winter and low in summer with narrow summer peaks. Such relatively high background concentrations are found at most eastern sites, indicating a year-round burning contribution.

The seasonal variation of ns-K concentrations allows us to segregate contributions from summer wildfires, other fires including prescribed fires, residential biofuel use, and industrial biofuel use. Wildfires are the most important contributor to ns-K concentrations in summer, especially in the west. Prescribed fires in the southeast generally do not take place in summer to avoid the high-ozone season. Residential biofuel use is negligible in summer and is the major contributor to the winter maximum in ns-K concentration, while industrial biofuel use is aseasonal (Cabada et al., 2002).

Fig. 2 shows the aseasonal background ns-K concentrations that we compute by averaging the lowest three monthly concentrations of individual years for 2001–2004. We attribute these to biofuel burning mostly by agro-industries since we expect all other types of biomass burning to be highly seasonal. Values are highest in the southeast and in northern Maine where the highest use of biofuel occurs in large pulp and paper mills (AF&PA, 2001; US EPA, 2002). We subtract these background values from the observed ns-K concentrations to estimate other burning contributions.

In summer, the observed ns-K concentrations in excess of the background values represent mostly wildfire contributions. There are fires in other seasons including wildfires and prescribed burns, which we classify as “other fires”. We differentiate

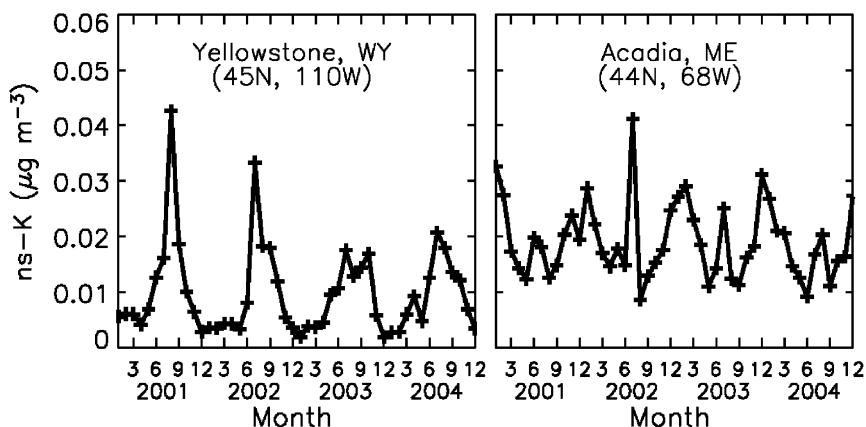


Fig. 1. Monthly mean non-soil potassium (ns-K) aerosol concentrations at Yellowstone (Wyoming) and Acadia (Maine) for 2001–2004.

contributions from these other fires from residential biofuel by using monthly gridded satellite-based dry mass burned data on a $2^\circ \times 2.5^\circ$ grid for 2001–2004 (van der Werf et al., 2006). The ns-K concentrations in excess of the background values in grid boxes with zero dry mass burned are taken to represent residential biofuel contributions. Some grid boxes with non-zero dry mass burned data may have biofuel contributions but in general we find a clear demarcation between the two sources, as conditions

propitious to fires are not associated with domestic heating needs.

The TC/ns-K emission ratio from biomass burning is highly variable depending on vegetation type and burning conditions (Andreae and Merlet, 2001). Extratropical forest fires generally have higher emission ratios ($20\text{--}130\text{ g C g}^{-1}$) than tropical forest or grassland fires ($10\text{--}20\text{ g C g}^{-1}$) (Andreae and Merlet, 2001). We estimate local fire emission ratios from the IMPROVE data as the slopes of TC vs. ns-K for individual sites and seasons with high correlations ($R > 0.7$, i.e., 50% of variance explained). The slopes are calculated using the reduced-major-axis method for linear regression (Hirsch and Gilroy, 1984).

Fig. 3 shows the slopes, i.e., the enhancement ratios $d[\text{TC}]/d[\text{ns-K}]$, for June–August for 2001–2004. Most sites with $R > 0.7$ are in the west; weaker fire influences and other TC aerosol sources (anthropogenic, biogenic) degrade the correlation in the east. Values range from 10 to $130\text{ }\mu\text{g C m}^{-3}/\mu\text{g m}^{-3}$. They are generally higher ($70\text{--}130\text{ }\mu\text{g C m}^{-3}/\mu\text{g m}^{-3}$) in the northern United States, where most of the burning is of forests, than in the south ($10\text{--}50\text{ }\mu\text{g C m}^{-3}/\mu\text{g m}^{-3}$), where burning of brush and grassland dominates (Bailey et al., 1994; Westerling et al., 2003). Wildfires in Canada contribute to high values in the northern United States, as discussed further below. The enhancement ratios vary year by year depending on burning conditions of fuels, in particular the

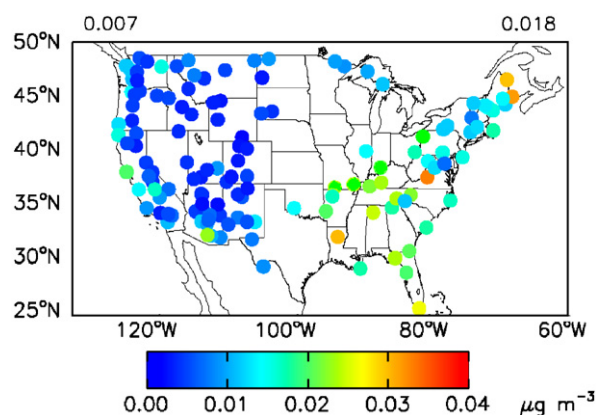


Fig. 2. Non-soil potassium (ns-K) aerosol background concentrations computed as means for 2001–2004 of the three lowest monthly average values of each year. Numbers above the plot give the mean concentrations in the western and eastern United States separated at 95°W .

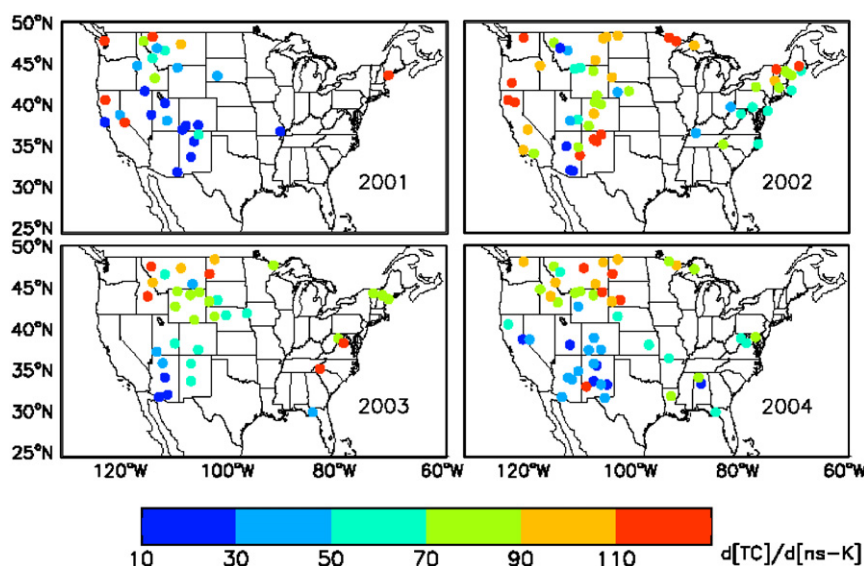


Fig. 3. Enhancement ratios ($\mu\text{g C m}^{-3}/\mu\text{g m}^{-3}$) of total carbonaceous (TC) to non-soil potassium (ns-K) aerosol at IMPROVE sites for June–August 2001–2004. The enhancement ratios are calculated as the slopes $d[\text{TC}]/d[\text{ns-K}]$ of the reduced-major axis linear regressions (Hirsch and Gilroy, 1984) for individual sites and years with strong correlation coefficients ($R > 0.7$) between the two species.

availability of moisture (Westerling et al., 2003). This interannual variability is most apparent in Colorado, Arizona, and New Mexico where values are highest in 2002, the warmest and driest summer on record (Waple and Lawrimore, 2003). The enhancement ratios from fires outside June–August (including prescribed burns) are also computed as described above and show a similar range to those for summer.

Deriving TC/K emission ratios from observed $d[TC]/d[ns-K]$ enhancement ratios works well for fires, which have large episodic sources, but not for biofuel use, where the source is more diffuse and the correlations are compromised by anthropogenic and biogenic sources of TC aerosol. We therefore adopt TC/ns-K biofuel emission ratios from the literature. Residential biofuel burning of different types of woods produces a fairly consistent potassium aerosol mass fraction of 1%, with TC contributing >90% of the aerosol (Fine et al., 2001), and so we use an emission ratio of 90. Industrial biofuel has a much lower emission ratio of 10 according to US EPA (2001), likely because it is burned more efficiently.

The emission ratios derived above are used to compute the burning contributions to TC aerosol concentrations at individual IMPROVE sites. For example, we derive the wildfire contributions to TC aerosol concentrations by multiplying the local $d[TC]/d[ns-K]$ enhancement ratios shown in Fig. 3 by the seasonal ns-K concentrations for that summer after subtracting background values from Fig. 2. For sites with $R < 0.7$, we apply the mean enhancement ratio averaged over the ensemble of sites with $R > 0.7$ within the same state. If no single site is available in the state for a particular year we use an average value over other years for that state. Other burning contributions are estimated similarly and details are discussed below.

3. Summer wildfire contributions

Here, we estimate the seasonal, annual, and interannual contributions of summer wildfires to TC aerosol concentrations for 2001–2004 using the method described in Section 2. Fig. 4 shows

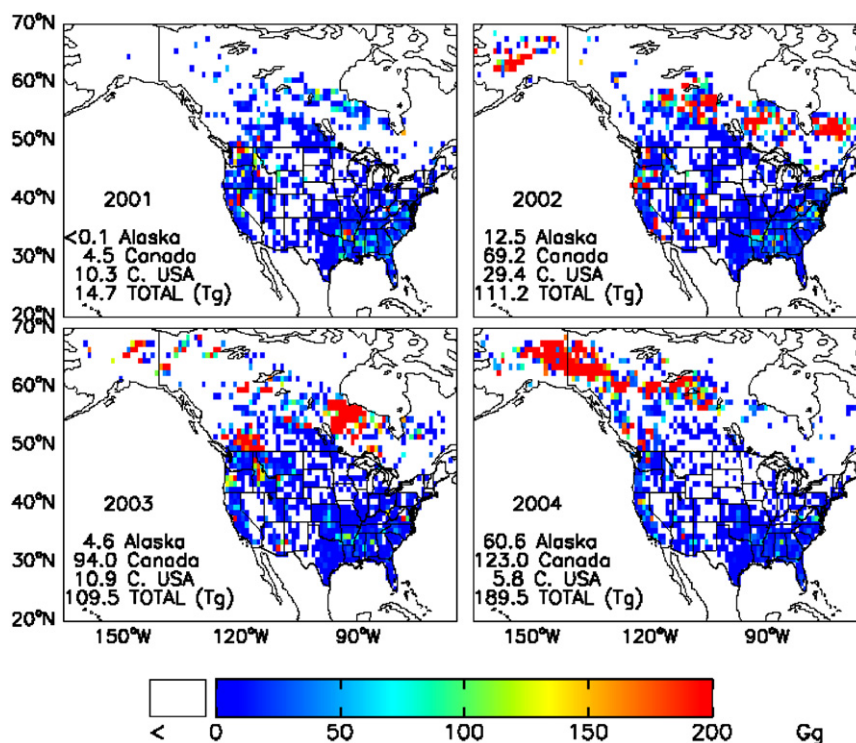


Fig. 4. Dry mass burned by fires in the United States and Canada in June–August 2001–2004 (van der Werf et al., 2006). Values are in units of Gg per $1^\circ \times 1^\circ$ grid square. Regional totals in unit of Tg are also given as inset for Alaska, Canada, and the contiguous United States of which the 8-yr averages for 1997–2004 are 12.6, 66.1, and 15.6 Tg, respectively.

seasonal dry mass burned in the contiguous United States, Alaska, and Canada for June–August 2001–2004 from van der Werf et al. (2006), who used satellite-derived monthly burned areas (Giglio et al., 2006) and fuel loads derived from the CASA biogeochemical model (Potter et al., 1993). The year-to-year variation in magnitude and patterns is mainly from wildfires. Fire emissions in the contiguous United States were highest in 2002 (double the 1997–2004 average), while 2004 was the largest wildfire season on record in Alaska (Turquety et al., 2007). The estimated dry mass burned in 2004 in Alaska is 61 Tg, a factor of 5 higher than the 8-yr average of 13 Tg. In Canada, the 2004 fire season was also well above the past 8-yr average, with 15 times the average area burned in the Yukon Territory (accounting for 60% of the national total) and six times the average area burned in British Columbia, according to the Canadian Interagency Forest Fire Center (CIFFC; Turquety et al., 2007).

Fig. 5 shows observed monthly mean TC concentrations averaged over all IMPROVE sites in the contiguous United States for 2001–2004. Values are highest in summer, due both to wildfires and to biogenic sources (Park et al., 2003). Wintertime concentrations, which are mainly from fuel use (Park et al., 2003), are highly reproducible from year to year. Summertime concentrations are much more variable and this can be largely explained by wildfires. Values are highest in 2002, corresponding to the wildfire maximum in the contiguous United States (Fig. 4). Interannual variability in biogenic emission is expected to be much smaller (Palmer et al., 2006). We find that the

interannual variability in Fig. 5 cannot be explained by temperature, indicating relatively small contributions of secondary organic aerosol to the variability. Although 2004 was the lowest wildfire year in the contiguous United States, the TC concentrations are relatively high in July when Alaskan and Canadian wildfires were most active. The long-range influence of boreal wildfires will be discussed in more detail below.

The estimated fire contributions to TC concentrations for June–August of 2001–2004 are shown in Fig. 6. Values are generally higher in the western United States than in the east. Highest values are found in California, Oregon, and Washington, where frequent wildfires occurred in every summer. The years 2002 and 2003 were particularly severe fire years in the west due to extreme hot and dry weather. In 2002, we find a maximum seasonal value of $5.2 \mu\text{g C m}^{-3}$ at Crater Lake National Park in Oregon (122.1°W , 42.9°N) and many west coast sites show values $>2 \mu\text{g C m}^{-3}$. Sites in Nevada, New Mexico, Colorado, and Arizona also show fire contributions exceeding $1 \mu\text{g C m}^{-3}$ in 2002. The high fire influence in the northwest in 2004 is due to long-range transport from fires in western Canada and Alaska (Fig. 4).

In the eastern United States, local wildfires are much less frequent than in the west. We still find high TC concentrations in the northeast (Fig. 6) due to transboundary transport from Canadian wildfires. This was the case in particular for fires in Quebec in 2002 (Colarco et al., 2004; DeBell et al., 2004). We find a maximum mean value of $3.2 \mu\text{g C m}^{-3}$ for June–August at Lye Brook Wilderness in Vermont (73.1°W , 43.1°N) which DeBell et al. (2004) previously identified as the IMPROVE site most affected by the Quebec fires. Remarkably, we find that fire contributions to TC concentrations in the northeast in 2003 and 2004 are similar to those in 2002, reflecting long-range transport from Canadian fires occurring further west than Quebec (Fig. 4).

Annual mean contributions to TC aerosol concentrations from summer wildfires averaged over 2001–2004 are shown in Fig. 7. As discussed above, the highest contribution is in the Pacific states with up to $0.7 \mu\text{g C m}^{-3}$ on an annual mean basis. Sites in Idaho, Montana, and Wyoming show annual contributions reaching $0.4 \mu\text{g C m}^{-3}$ due to both local fires and boreal wildfires in Alaska and Canada, particularly for 2004. The long-range transport of aerosols from boreal wildfires in

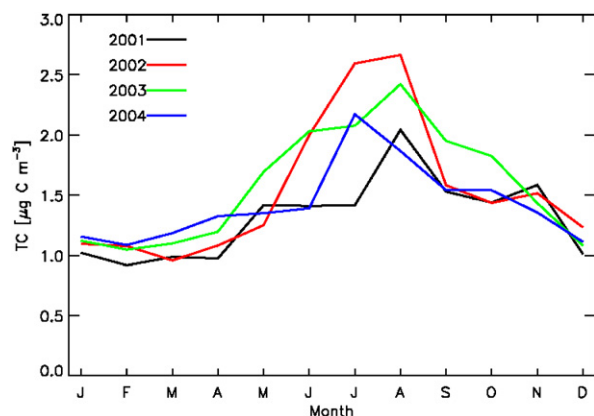


Fig. 5. Observed monthly mean concentrations of total carbonaceous (TC) aerosol in the contiguous United States for 2001–2004, averaged over all IMPROVE sites.

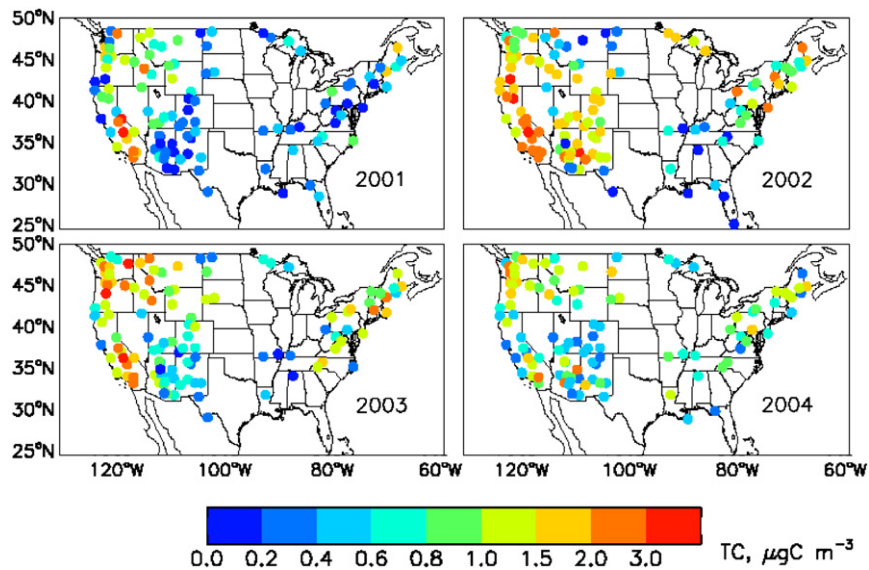


Fig. 6. Mean fire contributions to total carbonaceous (TC) aerosol concentrations in June–August 2001–2004.

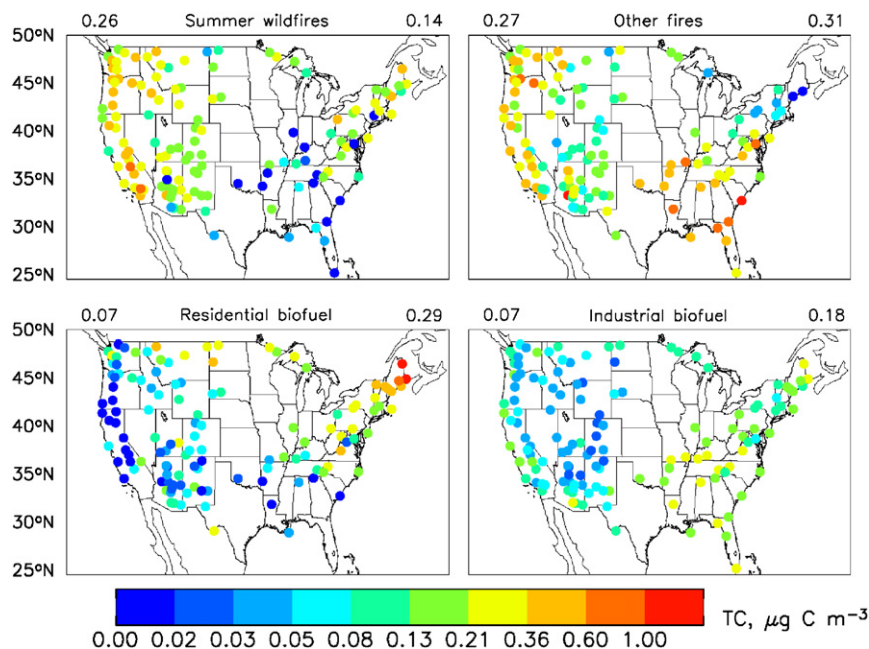


Fig. 7. Biomass burning contributions from summer wildfires, other open fires, residential biofuel, and industrial biofuel to annual mean total carbonaceous (TC) aerosol concentrations in the United States for 2001–2004. Numbers on top of each panel give the mean concentrations over the western and eastern United States separated at 95°W.

Canada also makes a significant contribution to TC aerosol concentrations in the northeast, with up to $0.4 \mu\text{gC m}^{-3}$ at sites in Maine. The regional annual mean TC concentrations due to summer wildfires for 2001–2004 are 0.26 and $0.14 \mu\text{gC m}^{-3}$ in the west and the east, respectively, and account for 22% and 7% of observed annual mean TC

concentrations (1.2 and $1.9 \mu\text{gC m}^{-3}$ in the west and east).

4. Other fires and biofuel contributions

Fig. 7 shows annual contributions of other fires, residential biofuel, and industrial biofuel to TC

aerosol concentrations in the United States. The contribution from other fires was obtained in the same manner as for summer wildfires but for the non-summer data. The contribution from residential and industrial biofuels was obtained by applying the TC/ns-K emission ratios given in Section 2. Other fires in the west represent mostly wildfires and agricultural fires. Highest contributions are in Oregon and Southern California, reaching $0.6 \mu\text{g C m}^{-3}$ on an annual mean basis. Other fires in the east are mostly prescribed burning especially in the southeast where the highest contributions amount to $1.0 \mu\text{g C m}^{-3}$ in South Carolina and Florida. The regional mean contributions from these non-summer fires to annual TC aerosol concentrations are 0.27 and $0.31 \mu\text{g C m}^{-3}$ for the west and the east, respectively.

Residential biofuel is extensively used for seasonal heating in rural areas of the northern United States (US EPA, 2001). The highest use is in the northeast because of cold winters and spring, an abundant wood supply, and a relatively large rural population. The highest concentration is $2.2 \mu\text{g C m}^{-3}$ at the Old Town Presque Isle site in Maine. The biofuel contribution in the west is relatively small except for North Dakota and part of Montana (up to $0.4 \mu\text{g C m}^{-3}$). Regional annual mean contributions are 0.07 and $0.29 \mu\text{g C m}^{-3}$ for the west and the east, respectively.

Industrial biofuels such as wood and agricultural residues are burned to provide energy and to remove waste products (US EPA, 2001). The highest use is in paper and pulp mills, which consume about 80% of industrial biofuels in the United States (Energy Information Administration, 2006) and are mostly in the southeast and Maine (AF&PA, 2001). Our estimates of industrial biofuel contributions to annual TC aerosol concentrations show values of up to $0.3 \mu\text{g C m}^{-3}$ in these regions. The annual mean TC concentrations due to industrial biofuels are 0.07 and $0.18 \mu\text{g C m}^{-3}$ in the west and the east, respectively.

5. Overall biomass burning contributions

We summarize in Table 1 our estimates of different biomass burning contributions to TC concentrations on an annual mean basis in the United States. Values are averaged over all IMPROVE sites and are separated into west and east; this separation is of interest for the Regional Haze Rule as discussed below. The total burning influence

Table 1

Biomass burning contributions to annual total carbon (TC) aerosol concentrations in the United States^a

	West	East
Summer wildfires	0.26	0.14
Other fires	0.27	0.31
Residential biofuel	0.07	0.29
Industrial biofuel	0.07	0.18
Total	0.67	0.92
Observed TC concentrations	1.24	1.90

^aConcentrations are in $\mu\text{g C m}^{-3}$. Values are annual and regional means averaged over IMPROVE sites in the contiguous United States for 2001–2004. West and east are separated at 95°W .

is 0.67 and $0.92 \mu\text{g C m}^{-3}$ for the west and the east, respectively, and in both regions accounts for about 50% of observed annual mean TC concentrations (1.2 and $1.9 \mu\text{g C m}^{-3}$ in the west and east). Most of the remainder is expected to be from fossil fuel combustion and vegetation sources (Park et al., 2003). Fires are the dominant biomass burning contribution in the west ($0.53 \mu\text{g C m}^{-3}$ vs. $0.14 \mu\text{g C m}^{-3}$ for biofuel) while biofuel is more important in the east ($0.47 \mu\text{g C m}^{-3}$ vs. $0.45 \mu\text{g C m}^{-3}$ for fires), and is dominant in the northeast. The fire influence is significant in the east, reflecting mainly prescribed fires in the southeast and Canadian fire plumes in the northeast.

In our previous study (Park et al., 2003), we fitted GEOS-Chem chemical transport model simulations of OC and EC aerosol contributions from different sources to the IMPROVE observations for 1998, and then generalized to other years using a climatological fire inventory (Duncan et al., 2003). We obtained TC aerosol contributions of 0.48 and $0.32 \mu\text{g C m}^{-3}$ from fires on an annual mean basis for the west and the east, respectively, consistent with our estimates (0.53 and $0.45 \mu\text{g C m}^{-3}$) shown in Table 1. The climatological fire TC emission in Park et al. (2003) for the contiguous United States was $0.68 \text{ Tg C yr}^{-1}$, while an independent inventory by Bond et al. (2004) estimated $0.89 \text{ Tg C yr}^{-1}$. This source range (0.7 – 0.9 Tg C yr^{-1}) seems consistent with the constraints from the IMPROVE observations.

The simulated annual TC concentrations from biofuel use in Park et al. (2003) were $0.21 \mu\text{g C m}^{-3}$ in the west and $0.93 \mu\text{g C m}^{-3}$ in the east, consistent with our estimate in the west ($0.14 \mu\text{g C m}^{-3}$) but

a factor of 2 higher in the east ($0.47 \mu\text{g C m}^{-3}$) where most of biofuel use takes place. Park et al. (2003) used a biofuel TC source of $0.96 \text{ Tg C yr}^{-1}$ for the contiguous United States, of which 80% was industrial. This differs from biofuel use emission inventories of $0.42 \text{ Tg C yr}^{-1}$ from Bond et al. (2004) and 0.42 Tg yr^{-1} PM_{2.5} from the US EPA 1999 National Emission Inventory (NEI99, <http://www.epa.gov/ttn/chief/>) with <10% from industrial biofuel. Park et al. (2003) used the same TC emission factors for industrial and residential biofuel burning, but that for industrial burning should be much lower as discussed in Section 2. Our constraints from the IMPROVE observations support the lower biofuel TC sources from Bond et al. (2004) and NEI99 but imply a higher (40%) contribution from industrial biofuel use (Table 1).

6. Implications for air quality standards

We now examine the implications of our results for air quality standards on annual mean fine aerosol mass concentrations in the United States. This requires converting the carbon mass derived above into total aerosol mass, to account for the non-carbon component of organic aerosol. A scaling factor of 1.4 has been commonly used (White and Roberts, 1977) but appears to be too low. Turpin and Lim (2001) recommend a value of 2.1 for non-urban air, and Malm et al. (2005) derived a value of 1.8 under heavy fire influence. We use 1.8 here.

Applying the factor of 1.8 to the estimated 90% TC present as OC, we derive annual mean biomass burning contributions to fine aerosol concentrations of 1.2 and $1.6 \mu\text{g m}^{-3}$ for the west and the east, respectively. The observed total fine aerosol mass concentrations including sulfate, ammonium, nitrate, soil dust, and carbonaceous aerosols average 3.8 and $8.2 \mu\text{g m}^{-3}$ at the IMPROVE sites in the west and the east for 2001–2004. Biomass burning accounts, respectively, for 30% and 20% of these totals, and represents a significant increment toward the health-based NAAQS for annual mean concentrations of fine aerosols ($15 \mu\text{g m}^{-3}$) (US EPA, 2004).

Protecting visibility is an additional regulatory concern in the United States. The US EPA Regional Haze Rule mandates a schedule of increasing emission controls for 2004–2018 to achieve linear improvement in visibility in US Federal Class I areas (national parks and other wilderness areas), at

a rate designed to meet an aspirational endpoint of natural visibility conditions by 2064. Natural visibility degradation is mainly from organic aerosol. US EPA (2003b) recommends default estimates for natural TC concentrations of $0.36 \mu\text{g C m}^{-3}$ in the west and $1.0 \mu\text{g C m}^{-3}$ in the east. We estimate annual fire influences of $0.53 \mu\text{g C m}^{-3}$ in the west and $0.45 \mu\text{g C m}^{-3}$ in the east. Vegetation makes comparable contributions of 0.47 and $0.55 \mu\text{g C m}^{-3}$ to natural TC in the west and the east, respectively (Park et al., 2003). Considering that the separation between anthropogenic and natural fires is difficult and to some extent arbitrary, the EPA-recommended default natural TC concentrations for application of the Regional Haze Rule appear reasonable.

7. Conclusions

Biomass burning is an important factor for air quality degradation in the United States on seasonal and annual time scales. We used observations of total carbonaceous (TC) aerosol concentrations and correlative information from the IMPROVE network of >100 sites nationwide to estimate and segregate the contributions from different types of biomass burning (summer wildfires, other fires, residential biofuel, and industrial biofuel) to total fine aerosol concentrations.

Our approach was to use non-soil potassium (ns-K) as a tracer of biomass burning, and to exploit the seasonal variation of ns-K as well as satellite observations of fires to separate contributions from different types of burning. The monthly mean ns-K concentrations in the west show strong summer peaks (wildfires) and low winter values, whereas in the east values are moderately high throughout the year and tend to peak in winter (residential biofuel).

We derived local TC/ns-K emission ratios for fires from sites with strong correlations between TC and ns-K concentrations. Values range from 10 to $130 \mu\text{g C m}^{-3}/\mu\text{g m}^{-3}$. They are highest in the northern United States (forest fires) and low in the south (brush and grassland fires), consistent with previous literature (Andreae and Merlet, 2001).

Multiplying the TC/ns-K emission ratios by the observed summertime ns-K concentrations in excess of background yielded the summer wildfire contributions to TC aerosol concentrations. Highest contributions are in the Pacific states, with a seasonal maximum of $5.2 \mu\text{g C m}^{-3}$ in Oregon in 2002. Although 2004 was a relatively low fire year in

the contiguous United States, we find a large fire influence in the northwest due to long-range transport from record wildfires in western Canada and Alaska. We also find significant wildfire influence in the northeast for most years, due to long-range transport from Canadian fires, resulting in seasonal TC concentrations as high as $3.2 \mu\text{g C m}^{-3}$ in Vermont in 2002.

We used MODIS satellite data to identify the locations of fires in seasons other than summer, and estimated the contribution from these fires (including wildfires and prescribed fires which) in a similar way using observed $d[\text{TC}]/d[\text{ns-K}]$ enhancement ratios. We find large contributions in the southeast (up to $1.0 \mu\text{g C m}^{-3}$ on an annual basis) due to prescribed burning.

We estimated the contribution from residential biofuel by using the remaining ns-K concentrations in excess of background, and the contribution from industrial biofuel on the basis of the ns-K background. The highest use of residential biofuel is in the northeast and the resulting annual TC concentration reaches $2.2 \mu\text{g C m}^{-3}$ in Maine. The contribution from industrial biofuel is mostly in the southeast where it reaches $0.3 \mu\text{g C m}^{-3}$ on an annual mean basis.

We thus estimate mean biomass burning contributions to annual TC aerosol concentrations in the United States of 0.67 and $0.92 \mu\text{g C m}^{-3}$ for the west and the east, respectively, accounting in both regions for about 50% of observed annual mean TC concentrations ($1.2 \mu\text{g C m}^{-3}$ in the west and $1.9 \mu\text{g C m}^{-3}$ in the east). Fires account for 80% of the biomass burning influence in the west (the rest is from biofuel) and 50% in the east. Fire influence in the east is mainly from prescribed fires in the southeast and Canadian fire plumes in the northeast.

Accounting for the estimated non-carbon fraction of the organic aerosol, we estimate annual mean biomass burning contributions to fine aerosol concentrations in the United States of 1.2 and $1.6 \mu\text{g m}^{-3}$ for the west and the east, accounting for 30% (west) and 20% (east) of the observed total fine aerosol concentrations. This represents a significant increment toward the health-based NAAQS for annual mean concentrations of fine aerosols ($15 \mu\text{g m}^{-3}$) (US EPA, 2004).

Our analyses support current bottom-up estimates of 0.7 – 0.9 Tg C yr^{-1} for the climatological (long-term average) source from open fires in the United States (Park et al., 2003; Bond et al., 2004).

They support the overall bottom-up biofuel emission inventories of $0.42 \text{ Tg C yr}^{-1}$ from Bond et al. (2004) and 0.42 Tg yr^{-1} PM_{2.5} from the US EPA NEI99 but ascribe a larger contribution from industrial biofuel (40%) than in these inventories ($<10\%$). The previous biofuel source estimate of 0.96 Tg yr^{-1} from Park et al. (2003) is too high because they assumed the same TC aerosol emission ratios for industrial and residential biofuel, whereas that for industrial biofuel is much lower.

Our analysis focused on 2001–2004, but the relative contribution of fires to aerosol concentrations will likely grow in the future because of continued decrease in US anthropogenic emissions (US EPA, 2004). Spracklen et al. (2007) show an increasing trend in OC aerosol concentrations in the western United States for 1989–2004, mostly driven by increases in wildfire activity linked to warmer spring and summer temperatures (Westerling et al., 2006). Continuing increases in wildfires as a result of climate warming and accumulated stocks may have significant impact on future air quality.

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