Atmospheric Environment 45 (2011) 5407–5414

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

$PM_{2.5}$ source profiles for black and organic carbon emission inventories

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ARTICLE INFO

Article history:
Received 1 April 2011
Received in revised form
30 June 2011
Accepted 5 July 2011

Keywords:
Source profile
Emission inventory
$PM_{2.5}$
Black carbon
Elemental carbon
Organic carbon
Climate

ABSTRACT

Emission inventories for black or elemental (BC or EC) and organic (OC) carbon can be derived by multiplying $PM_{2.5}$ emission estimates by mass fractions of these species in representative source profiles. This study examines the variability of source profiles and its effect on EC emission estimates. An examination of available profiles shows that EC and OC ranged from 6–13% and 35–40% for agricultural burning, 4–33% and 22–68% for residential wood combustion, 6–38% and 24–75% for on-road gasoline vehicles, and 33–74% and 20–47% for on-road heavy-duty diesel vehicles, respectively. Source profiles from the U.S. EPA SPECIATE data base were applied to $PM_{2.5}$ emissions from the U.S. EPA National Emissions Inventory for 2005. The total estimated EC emissions of 432 Gg yr^{-1} was apportioned as 42.5% from biomass burning, 35.4% from non-road mobile sources, 15% from on-road mobile sources, 5.4% from fossil fuel (e.g., coal, oil, and natural gas) combustion in stationary sources, 1% from other stationary industrial sources, and 0.5% from fugitive dust. Considering the variability in available source profiles, BC emission estimates for major sources such as open fires and non-road diesels ranged from 42 to 133 (a factor of 3) and 25 to 100 (a factor of 4) Gg yr^{-1}, respectively. The choice of source profiles can be a major source of uncertainty in national and global BC/EC emission inventories.

1. Introduction

The global direct radiative effect of light-scattering particulate matter (PM) containing sulfates and nitrates is on the order of −1 Watts per square meter (W m^{-2}) and causes global cooling. PM containing black carbon (BC; also called elemental carbon [EC], light-absorbing carbon, or "soot") from fossil fuel and biomass combustion, produces a warm forcing of +0.2 to +1 W m^{-2} (IPCC, 2007). Combustion processes also produce organic carbon (OC) that affects radiative forcing mainly through light scattering. BC rarely occurs in the absence of OC, however, and light absorption efficiencies depend on the compositions, shapes, and sizes of BC particles (Fuller et al., 1999). Accurate BC and OC emission inventories are needed for estimating the effects of aerosols on climate change.

The U.S. EPA National Emissions Inventory (NEI; U.S. EPA, 2011a) includes annualized emission rates for $PM_{2.5}$ and PM_{10} (particles with aerodynamic diameters < 2.5 and 10 µm, respectively). Chemically-speciated emission rates can be estimated by multiplying PM emission rates from similar sources by source profile abundances of chemical species such as BC and OC (Reff et al., 2009; Chow et al., 2010a). PM source profiles are determined by real-world emission tests on representative source-types followed by chemical speciation (Chow and Watson, in press). Composite profiles average chemical abundances from several tests of similar sources (e.g., Chow et al., 2003) using the standard deviation as an estimate of uncertainty. Hundreds of individual source profiles and many composites with BC and OC (as carbon mass) $PM_{2.5}$ mass fractions (abundances) have been compiled in the U.S. EPA's (2008) SPECIATE Version 4.2 data base. These profiles represent source categories such as: 1) Power Plant, Incinerator, and Industrial Stationary sources; 2) Agricultural Burning, Open Burning, Forest Fire, and Residential and Industrial Wood Combustion Area sources; and 3) On- and Non-road Mobile sources. The objective of this study is to examine BC and OC abundances in source profiles from SPECIATE along with additional profiles obtained by the authors (Chang et al., 2004; Chow et al., 1992, 2003, 2004; England et al., 2007; Fujita et al., 2007a,b; Green et al., 2004; Kuhns et al., 2004; Mazzera et al., 2001; Watson and Chow, 2001; Watson et al., 1994, 2001, 2008a; Zielinska et al., 2008) to evaluate their variability within

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doi:10.1016/j.atmosenv.2011.07.011

and between source-types and to assess the effect of this variability on BC and OC annual U.S. emission rates.

2. Methods

The emission of any species from a particular source is the product of an emission factor and an activity for that source. Emission factors have been measured directly or estimated with models such as the MOtor Vehicle Emission Simulator (MOVES, U.S. EPA, 2011b). Activities have been estimated from fuel use for stationary and area sources, vehicle miles traveled (VMT) for mobile sources, or surrogates such as population. Thus, BC or OC emissions from source k may be calculated according to Equation (1):

$$BC_k = A_k EF_k (1 - eff_m) X_{k,m} F_k \tag{1}$$

where BC_k, emissions of BC (or OC) for source k; A_k, activity for source k (e.g., coal consumption in power plants or VMT for diesel trucks); EF_k, emission factor for PM from source k (e.g., in units of grams per VMT or unit fuel consumed); eff_m, emission reduction efficiency of the emission control technology m and; $X_{k,m}$, actual implementation rate of the emission control measure for source k and emission control technology m (e.g., percent of total coal combusted in power plants that are equipped with electrostatic precipitators); F_k, BC (or OC) fractional abundance in PM from source k.

If no emission controls are applied, the abatement efficiency equals zero (i.e., $eff = 0$) and the implementation rate is one (i.e., $X = 1$). In that case, the emission calculation is reduced to the product of the activity and the emission factor. PM ($PM_{2.5}$ and PM_{10}) emissions (accounting for all terms on the right side of Equation (1) except F_k) from thousands of sources are integrated spatially and temporally in the U.S. EPA NEI. Thus, BC and OC emissions can be calculated for each source in the NEI by applying an appropriate source profile to the PM emissions.

3. Results and discussion

3.1. Variability of BC and OC source profiles

Since source profiles are based on chemical, not optical, measurements, their BC content is discussed in terms of EC abundance, which is highly correlated with BC (Chow et al., 2010b), but the two are not necessarily equal. The variability of emission composition may be quantified by averaging chemical mass fractions of $PM_{2.5}$ from similar sources (Chow et al., 2003) and using the standard deviation to estimate the uncertainty. Supplemental Table S-1 lists 109 $PM_{2.5}$ source profiles for EC and OC (mostly composites unless otherwise noted) representing source categories including: fossil fuel combustion (e.g., coal, oil, and natural gas); other industrial stationary sources; biomass burning; fugitive dust; and on- and non-road mobile emissions. These profiles were selected because they: 1) have complete PM speciation (i.e., elements, ions, EC, and OC); 2) are relevant to current source types and pollution controls; 3) correspond to source categories in the U.S. NEI (U.S. EPA, 2011a); 4) have reasonable quality ratings (U.S. EPA, 2008); and 5) the original reports and publications describing the data could be obtained and reviewed (for the most part). The U.S. EPA assigns the highest quality ratings to the most recently determined profiles estimated from the largest number of samples (U.S. EPA, 2008).

Owing to well-controlled combustion conditions, modern U.S. industrial emissions do not contain large EC abundances (<5%), as shown in Fig. 1. In many of these profiles, EC was below the minimum detectable limit and the reported OC may contain organic vapors adsorbed onto the quartz-fiber filter (Chow et al., 2010c; Watson et al., 2009). The low $PM_{2.5}$ carbon content of these emissions may reflect the effectiveness of emission controls and/or be due to the high-temperature combustion in these industrial processes. As shown in Supplemental Table S-1, sawdust and phosphate industries (SPECIATE profile IDs 222022.5 and

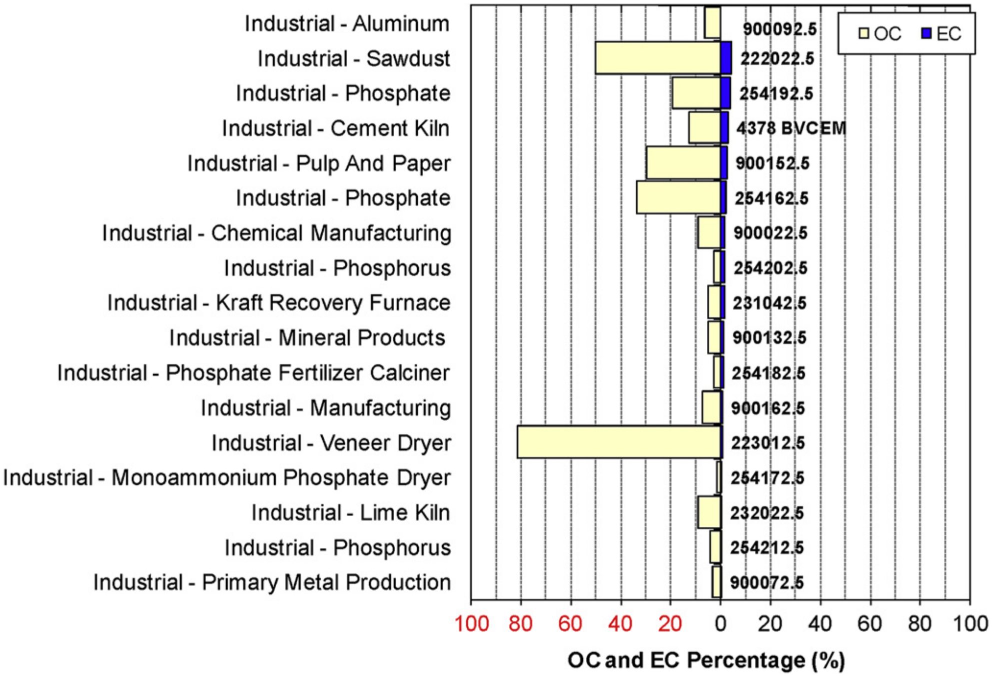

Fig. 1. EC and OC abundances in selected composite $PM_{2.5}$ stationary industrial source profiles other than fossil fuel combustion. U.S. EPA SPECIATE source profile ID numbers and original mnemonics are noted to the right of the EC bar and detailed in Supplemental Table S-1.

254192.5, respectively) show the highest $PM_{2.5}$ EC abundances at 4–5%, followed by cement kiln (3.0 ± 2.5% in BVCEM; Chow et al., 2004) and pulp and paper manufacturing (2.6 ± 1.8% in SPECIATE profile ID 900152.5).

Fig. 2 displays $PM_{2.5}$ EC and OC abundances from biomass burning emissions. Excluding one outlier (SPECIATE profile ID 422012.5 with 33 ± 13% EC), the EC abundance varies over fivefold, ranging from 3.2 ± 1.8% (MZFFIREC; Northwestern Colorado forest fire; Watson et al., 1996) to 17.5 ± 1.6% (NWFGPDa; Denver residential wood combustion-fireplace; Zielinska et al., 1998). The OC variability is smaller, within a factor of two to three, from 22.3 ± 13.6% (LTWOODST; Lake Tahoe woodstove; Kuhns et al., 2004) to 67.6 ± 5.9% (NWFGPDa; Zielinska et al., 1998). This is because OC is the major component of $PM_{2.5}$ in the profiles in Fig. 2.

In Fig. 3, $PM_{2.5}$ EC abundances for gasoline engine exhaust range from 5.9% (SPECIATE profile ID 312302.5, southern California; Cass and McRae, 1981) to 37.5 ± 8.5% (NWLCPC, low emitter; Denver, CO; Zielinska et al., 1998). $PM_{2.5}$ EC abundances for gasoline engines are generally 20–40% lower than those reported for diesel engines (Fig. 4). Fujita et al. (2007a) found a subset of gasoline-fueled vehicles that reported high EC emissions under cold (SI_BC; 37.4 ± 10.6% EC) and warm start (SI_BW; 34.6 ± 12.6% EC) conditions as shown in Supplemental Table S-1. Cold and warm start conditions do not appear to influence EC or OC abundances (31–32% OC) in the gasoline-fueled engine exhaust profiles. As shown in Fig. 3, $PM_{2.5}$ EC abundances in a wintertime gasoline low-emitter profile (NWLCPC; Zielinska et al., 1998) are similar to those of the SI_BC and SI_BW profiles. High emitter or "smoker" (NWHCPC; 34.0 ± 20.3%) and low-emitter or "non-smoker" (NWnSPC; 35.9 ± 12.8%) winter profiles also reported similar EC abundances. For summertime high-emitter vehicles (e.g., NSSCPC), the fraction of EC decreases to 9.2 ± 8.7% while the fraction of OC increases to 75.0 ± 7.3%); the excess emissions are therefore dominated by OC.

Fig. 4 shows EC abundances from on-road heavy-duty diesel engine exhaust range from 32.9 ± 8.0% (PHDIES) to 73.8 ± 28.2% (HDD) of $PM_{2.5}$ mass. EC and OC in the PHDIES (Watson et al., 1994) and HDD (Fujita et al., 2007a,b) profiles were measured with the same analytical method (i.e., the IMPROVE protocol with thermal/optical reflectance [TOR] pyrolysis correction; Chow et al., 1993, 2007, 2011) but the two studies were separated by 12 years. PHDIES represents average diesel-fueled vehicle emissions sampled at a centralized inspection and maintenance facility in Phoenix, Arizona during winter 1989, while the HDD profile was determined from dynamometer tests in southern California during summer 2001 following Federal Test Procedure (FTP)-compatible test cycles. This over twofold variation in EC abundances may reflect differences in operating conditions during the tests and implementation of emission controls over the last two decades. The higher EC abundance in the HDD profile may better represent emissions from current on-road heavy-duty diesel fleets.

The EC abundance from the On-road Heavy-duty Diesel engine vehicle profile (NWHDc; 73.5 ± 10.1%) (Watson et al., 1998; Zielinska et al., 1998) is similar to the HDD profile (73.8 ± 28.2%), but the OC fraction differs by over twofold (19.8 ± 7.7% in NWHDc versus 47.2 ± 18.7% in HDD). OC abundances can differ owing to engine operating conditions at high altitude in Denver, Colorado for the NWHDc profile (Watson et al., 1990, 1998) and by adsorption of volatile organic compounds onto quartz-fiber filters during sampling (positive artifact; Turpin et al., 1994; Watson and Chow, 2002; Chow et al., 2006, 2008, 2010c; Watson et al., 2008b, 2009). In Fujita et al. (2007a,b), OC, EC, and $PM_{2.5}$ were normalized to total carbon concentrations while in this study they were normalized to $PM_{2.5}$ concentration.

For light-duty and medium-duty diesel engines, EC abundances range from 61–76%, similar to those for on-road heavy-duty diesel engines. For the non-road composite profile (LVOffRDIE; Green et al., 2004), the EC abundance (23.7 ± 11.9%) is ∼50% lower compared to the on-road diesel profiles of 45.4 ± 12.9% (LVOnRDIE)

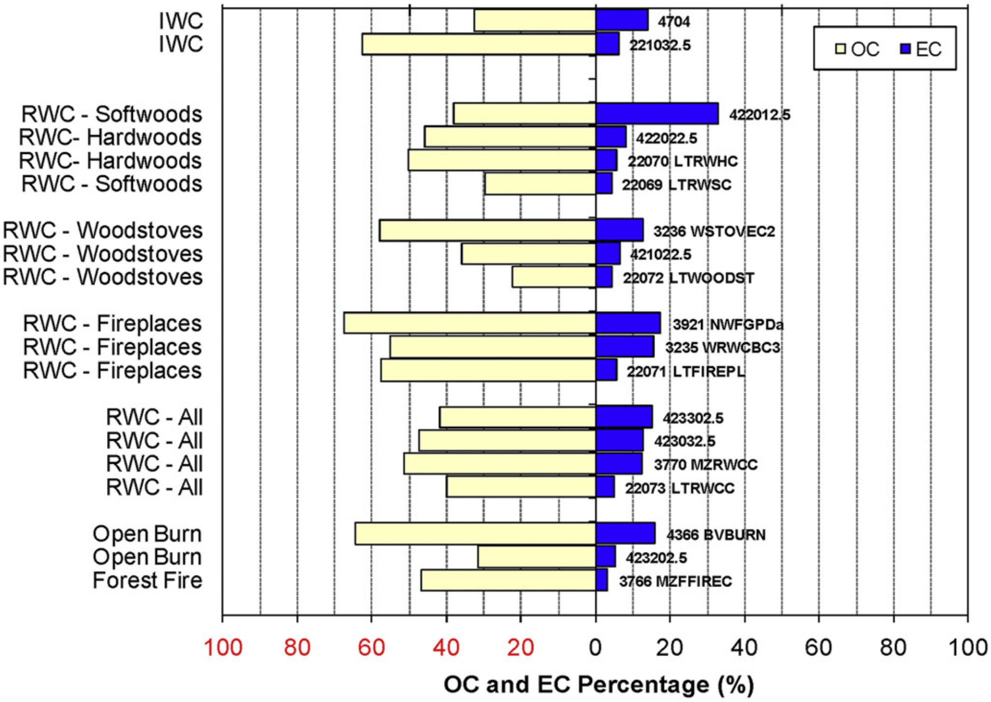

Fig. 2. EC and OC abundances in selected composite $PM_{2.5}$ source profiles representing wood burning emissions. IWC = Industrial Wood Combustion and RWC = Residential Wood Combustion. U.S. EPA SPECIATE source profile ID numbers and original mnemonics are noted to the right of the EC bar and detailed in Supplemental Table S-1.

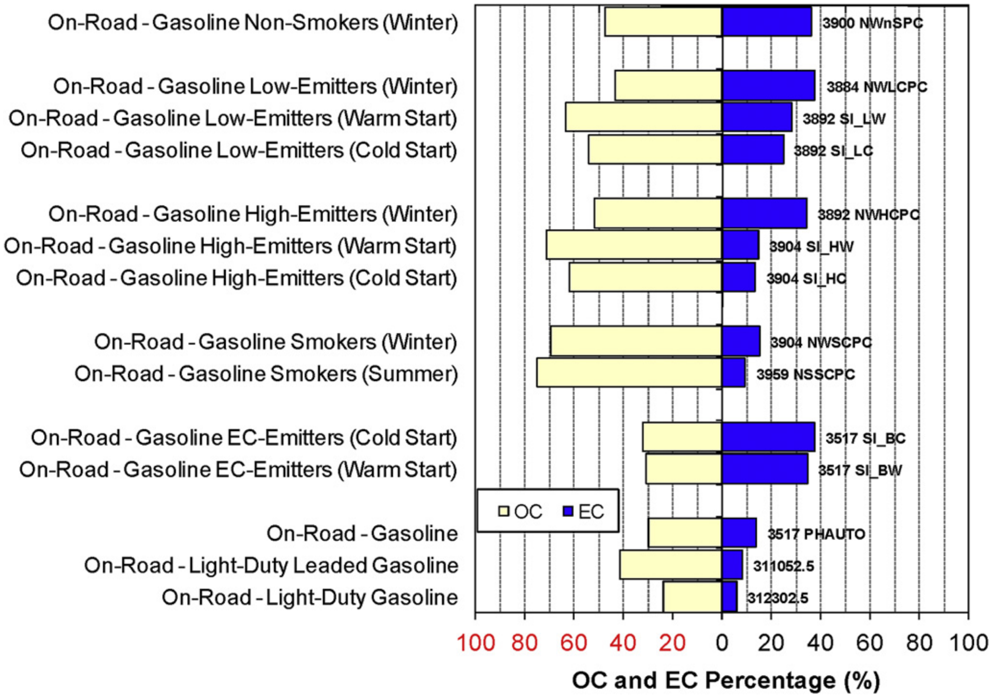

Fig. 3. EC and OC abundances in selected composite $PM_{2.5}$ source profiles representing gasoline-fueled vehicle emissions. U.S. EPA SPECIATE source profile ID numbers and original mnemonics are noted to the right of the EC bar and detailed in Supplemental Table S-1.

to 58.3 ± 28.2% (LVOnRDIEs) during the winter and summer, respectively, in Las Vegas, Nevada. On the other hand, the non-road PEN_C profile from Military Diesel Generators (Watson et al., 2008a) shows an EC abundance (15.3 ± 9.2%) more comparable to that of the non-road LVOffRDIE profile.

Different fuels, equipment, pollution controls, operating conditions, and test methods cause variability in EC and OC abundances, and these are not fully specified in U.S. EPA (2008), or even in the original references (Supplemental Table S-1). Another cause of variability is the difference in the OC/EC split for different carbon measurement methods (Watson et al., 2005). Currie et al. (2002) found EC concentration differences up to a factor of seven among different laboratories analyzing the same samples.

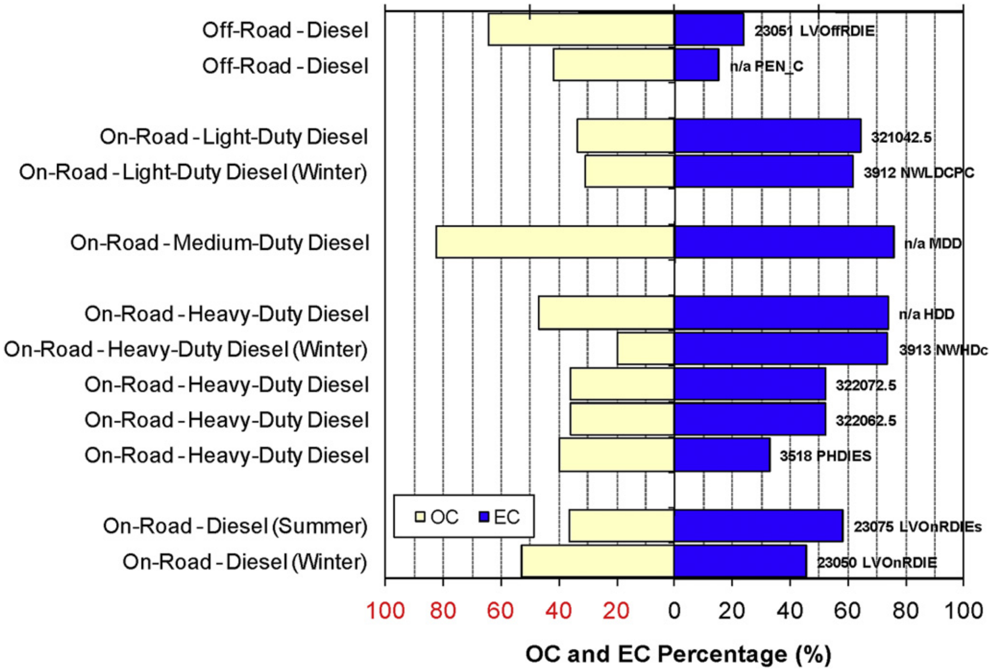

Fig. 4. EC and OC abundances in selected composite $PM_{2.5}$ source profiles representing diesel engine exhaust emissions. SPECIATE profile numbers and original mnemonic are noted to the right of the EC bar and detailed in Table S-1. SPECIATE profile ID numbers 322072.5 and 322062.5 reported identical OC and EC abundances with different $PM_{2.5}$ sum of species [89.06 and 88.71%, respectively] and have different references in U.S. EPA (2008).

3.2. Comparisons of source profiles from independent compilations

Reff et al. (2009) constructed chemically-speciated emission rates based on the 2001 Clean Air Interstate Rule (CAIR) version of the NEI (http://www.epa.gov/air/interstateairquality/technical.html) using composite and simplified profiles shown in Supplemental Table S-2, prefaced with SPECIATE Version 4.2 codes in the 91000 and 92000 series, respectively. Reff et al. chose the median of SPECIATE profiles for a given source category. Uncertainties for the simplified and composite source profiles are not provided in SPECIATE, as there is no statistically meaningful basis for assigning an uncertainty to a median value. Values and ranges of $PM_{2.5}$ EC and OC abundances from Supplemental Table S-1 are compared with the Reff et al. (2009) values in Table 1 using identical EC and OC source values for the Industrial sources not categorized by specific fuels (i.e., Manufacturing, Mineral Products, Kraft Recovery Furnace, Chemical Manufacturing, Pulp and Paper, and Cement Kiln). OC and EC abundances are similar, but not identical, for the Incinerator, Fly Ash, Residential Coal Combustion, and Charbroiling categories. They fall within ranges of abundances for Agricultural Burn, Residential Wood Combustion, and On-road Light-duty Gasoline vehicle categories.

The Residential Natural Gas Combustion profile (ID 92063) was measured by Hildemann et al. (1991), but Reff et al. (2009) set the EC abundance to zero due to the sum of fractional abundances being greater than one in the original profile (SPECIATE profile ID 421072.5). In both cases, the profiles are dominated by OC (83–85%). The Natural Gas Boiler profile reported by England et al. (2007) contains 13 ± 11% EC and 61 ± 23% OC.

For wildfires, profile ID 92090 contains a 9.5% EC abundance, which is an average of the forest fire in northern Colorado (NZFFIREC; 3.2 ± 1.8% EC; Watson et al., 1998) and an open fire in western Texas (BVBURN; 15.8 ± 14.5% EC; Chow et al., 2004). It is now known that EC abundances are higher during the flaming phase than the smoldering phase of biomass burning (Chen et al., 2007) and are influenced by fuel moisture content (Chen et al., 2010). Light absorbing properties in the smoldering phase depend strongly on the wavelength of light (Chen et al., 2006; Kirchstetter et al., 2004; Park et al., 2006).

3.3. Elemental and organic carbon emission inventories

The 2005 NEI (U.S. EPA, 2011a) contains 4270 source categories characterized at Source Classification Code (SCC) 4, the most detailed level. A subset of 3794 SCC4 categories account for 99.4% of U.S. $PM_{2.5}$ emissions, and these can be grouped into 90 categories (based on the SCC descriptions) as shown in Supplemental Table S-2, believed to have similar EC and OC abundances (U.S. EPA, 2008). Supplemental Table S-3 tabulates the corresponding EC and OC annual emission rates obtained by multiplying the $PM_{2.5}$ mass emissions by the EC and OC abundances.

All of the emissions in U.S. EPA (2011a) are traceable to submissions from the states except for the category of Open Fires (SCC 2810090000). These emissions apparently result from a separate nationwide fire survey by satellite remote sensing (Pouliot et al., 2008). As such, there is a potential for double-counting of large fires such as those included in the Agriculture Burning, Open Burning, Wildfires, Slash Burning, and Prescribed

Table 1
Comparison of $PM_{2.5}$ EC and OC abundances for selected source categories from two independent compilations.

Source category	EC (%)		OC (%)	
	This study[a]	SPECIATE 4.2[b]	This study	SPECIATE 4.2
I. Stationary sources				
Coal-fired power plants	1.38–4.1	0.13–1.97	5.2–27	2.6–39
Oil combustion	3.0–13.6	1.0[c]	2.0–5.0	1.0[a]
Gas-fired boilers[d]	13	38	61	25
Gas-fired process heaters[d]	6.3	17.2	62	35
Incinerator	2.4	1.52	13.4	8.4
Fly ash	1.39	1.70	1.53	1.24
Industrial – lime kiln	0.46	2.3	9.3	6.6
Industrial – manufacturing	**0.89**	**0.89**	**7.4**	**7.4**
Industrial – mineral products	**1.47**	**1.47**	**5.2**	**5.2**
Industrial – kraft recovery furnace	**1.53**	**1.53**	**5.2**	**5.2**
Industrial – chemical manufacturing	**1.83**	**1.83**	**9.2**	**9.2**
Industrial – pulp and paper	**2.6**	**2.6**	**30**	**30**
Industrial – cement kiln	**3.0**	**3.0**	**12.8**	**12.8**
II. Area sources				
Agricultural burn	5.8–13.1	10.9	34–40	39
Forest fire	3.2	9.5[e]	47	56[c]
Residential wood combustion	4.2–33	5.6[f]	22–58	53[f]
Residential coal combustion	26	24	70	64
Residential natural gas combustion	6.7	0	85	83
Charbroiling	3.6	4.1	95	81
III. Mobile sources				
On-road – light-duty gasoline	5.9–37	21	24–75	58
On-road – heavy-duty diesel	33–74	77	19.8–47	17.6
On-road – light-duty diesel	62–64	58	31–34	40
Tire wear	29–61	22	22–58	47

[a] From this study, see Supplemental Table S-1.
[b] See Supplemental Table S-2.
[c] Residual oil.
[d] The data were from the same study. This study reference is England et al. (2007). SPECIATE reference is Chang et al. (2004).
[e] Average of Forest Fire (SPECIATE profile ID 3766; MZFFIREC) and Open Burn (SPECIATE profile ID 4366; BVBURN) in Supplemental Table S-1, referred to by Reff et al. (2009) as “Wildfires” in Supplemental Table S-2.
[f] SPECIATE profile ID 92068; composite of hardwood and softwood.

Burning categories. As shown in Supplemental Table S-3, Open Fire emissions exceed the sum of other large fire categories (63.6 Gg yr^{-1}) by 27.3 Gg yr^{-1} for EC and 209 Gg yr^{-1} for OC. The Open Fire category accounts for 50% of total biomass burning EC emissions and 21% of total U.S. annual EC emissions. Open Fires are treated here as a separate category, recognizing that there may be some double counting in the 2005 NEI. This study used the Prescribed Burning profile (SPECIATE profile ID 92059; Supplemental Table S-2) to represent Open Fires and Open Burning sources, with an EC abundance of 10.93% (within the range of 5–16% for the Open Burning source in Supplemental Table S-1). Accounting for profile variability, EC emissions for the Open Fire source (90.9 Gg yr^{-1}) in Supplemental Table S-3 could range from to 42 to 133 Gg yr^{-1}. The heavy-duty diesel profile (HDDV; EC = 77.1%, OC = 17.6%; SPECIATE profile ID 92035; Supplemental Table S-2) was chosen to represent all non-road diesel sources because this category includes heavy mining and agricultural equipment and vehicles (e.g., tractors, backhoes, loaders, excavators, trucks, and combines), similar to on-road heavy-duty diesel trucks. However, the average EC content in the two non-road diesel profiles (fork lift and generator) in Supplemental Table S-1 is only 19.5%. Using this value, EC emissions from non-road diesel sources would decrease by a factor of four, from 100 to 25 Gg yr^{-1}. The range of estimates for EC emissions from open fires and non-road diesel sources based on different source profiles is much larger than uncertainties assigned to the corresponding source profiles (Table S-1).

Using the selected source profiles for 2005 emissions (Supplemental Table S-2), the breakdown of the 432 Gg yr^{-1} EC emissions results in 5.4% from fossil fuel (e.g., coal, oil, and natural gas) combustion in stationary sources; 1% from other stationary industrial sources; 42.5% from biomass burning; 0.5% from fugitive dust; 15% from on-road mobile sources; and 35.4% from non-road mobile sources. Of the 1395 Gg yr^{-1} 2005 OC emissions, the breakdown results in 5.6% from fossil fuel combustion in stationary sources; 2% from other stationary industrial sources; 80% from biomass burning; 5.5% from fugitive dust; 2.3% from on-road mobile sources; and 5% from non-road mobile sources.

Streets et al. (2004) report U.S. 1996 EC emissions of 414 Gg yr^{-1} derived from Bond et al. (2004), similar to the 432 Gg yr^{-1} for the 2005 NEI. Streets et al. (2004) categorized emissions into Industry, Power, Residential, Biomass Burning, and Transport categories. Transport EC emissions of 203 Gg yr^{-1} are consistent with the sum of 2005 Mobile on-road and non-road EC emissions of 219 Gg yr^{-1}. However, the 1996 industrial EC emissions of 66 Gg yr^{-1} (Streets et al., 2004) are over two times higher than the sum of all fossil fuel combustion (23.1 Gg yr^{-1}) and other industrial emissions (4.4 Gg yr^{-1}) in Supplemental Table S-3.

Battye and Boyer (2002) estimated U.S. emissions from the 1999 NEI using an earlier version of the SPECIATE data base, and these are compared with the 2005 estimates in Table 2. The total EC emissions of 433 and 432 Gg yr $^{-1}$ in the 1999 and 2005 inventories, respectively, are nearly identical. However, contributions from source categories differ, as compared for the large non-stationary source categories in Table 2. The differences in the Unpaved Road Dust EC source profiles (1% in 1999 versus 0.1% in 2005) led to a large EC emission difference (i.e., 13 Gg yr^{-1} in 1999 versus 0.7 Gg yr^{-1} in 2005). The 1% EC composition in the 1999 Unpaved Road profile appears too large, considering that the average EC composition in all Unpaved Road Dust profiles in SPECIATE Version 4.2 is 0.3% ($N = 86$). On-road and non-road Diesel EC emissions were similar for 1999 and 2005 (65 and 91 Gg yr^{-1}, respectively, in 1999, and 59 and 100 Gg yr^{-1}, respectively, in 2005) because PM$_{2.5}$ emissions from these sources in 1999 (151 and 211 Gg yr^{-1}, respectively) were nearly double those in 2005 (77 and 130 Gg yr^{-1}, respectively) while the PM$_{2.5}$ diesel EC abundance for 2005 (77%) was nearly double that in 1999 (43%). PM$_{2.5}$ emissions for the Locomotive Diesel source were nearly the same in 1999 (25 Gg yr^{-1}) and 2005 (26 Gg yr^{-1}) but the EC emissions in 2005 (20 Gg yr^{-1}) were nearly double those in 1999 (11 Gg yr^{-1}) because of the respective differences in the EC source profiles.

Table 2
Comparison of area and mobile EC emissions based on the 1999[a] and 2005[b] NEI PM$_{2.5}$ inventories.

	Emission rate (Gg yr^{-1})				Percentage PM$_{2.5}$ mass	
	1999	2005	1999	2005	1999	2005
	PM$_{2.5}$	PM$_{2.5}$	EC	EC	EC[c]	EC[c]
II. Area sources						
Agricultural burning	85	204	10	22	12	11
Wildfires	212	161	15	15	7.2	9.5
Residential wood combustion	340	347	21	19	6.1	5.6
Unpaved road dust	1283	764	13	0.7	1	0.1
Paved road dust	620	111	10	1.2	1.7	1
III. Mobile sources						
Non-road diesel	211	130	91	100	43	77
On-road diesel	151	77	65	59	43	77
Non-road gasoline	75	50	20	5	27	10
On-road gasoline	58	22	16	4.6	27	21
Aircraft	25	9	17	6.9	70	76
Locomotive diesel	25	26	11	20	43	77

[a] Battye and Boyer (2002).
[b] Current study based on U.S. EPA (2011a,b).
[c] EC source profile.

4. Conclusions

There is a wide range of EC and OC abundances in PM$_{2.5}$ source profiles representing the same source type. For profiles compiled in the U.S. EPA's SPECIATE library, EC and OC ranged from 6–13% and 35–40% for Agricultural Burning, 4–33% and 22–68% for Residential Wood Combustion, 6–38% and 24–75% for On-road Gasoline vehicles, and 33–74% and 20–47% for On-road Heavy-duty Diesel vehicles, respectively. The U.S. EPA National Emissions Inventory (NEI) for 2005 provides emission estimates for PM$_{2.5}$ from thousands of stationary, area, and mobile sources. Applying the source profiles to the corresponding PM$_{2.5}$ emissions, total U.S. EC and OC emissions were 432 and 1395 Gg yr^{-1}, respectively. The largest EC emitters were Non-road Diesel (100 Gg yr^{-1}), Open Fires (91 Gg yr^{-1}; from satellite measurements), and On-road Diesel (59 Gg yr^{-1}). The largest OC emitters were Open Fires (533 Gg yr^{-1}), Residential Wood Combustion (183 Gg yr^{-1}), and Open Burning (144 Gg yr^{-1}).

Estimated EC emissions are sensitive to the choice of source profile. EC emissions from Open Fires (91 Gg yr^{-1}) could range from 42 to 133 Gg yr^{-1} based on the variation in source profiles. A Heavy-duty Diesel vehicle source profile with an EC composition of 77% was used to represent all non-road diesel emissions, but non-road diesel EC abundances are as low as 19.5% in published profiles. Based on the smaller value, non-road diesel EC emissions would be lower by a factor of four, i.e., 100 versus 25 Gg yr^{-1}. Using the "lower-limit" source profiles for just these two sources would decrease the total EC emissions by 120 Gg yr^{-1} (28%). Our EC emissions estimate of 432 Gg yr^{-1} is consistent with those reported for earlier periods (433 Gg EC yr^{-1} for 1999; Battye and Boyer, 2002; and 414 Gg EC yr^{-1} for 1996; Streets et al., 2004). This agreement was fortuitous for some sources where differences in source profiles were offset by opposite differences in PM emissions.

While the U.S. EPA SPECIATE data base is a comprehensive compilation, there is no universal consensus on the choice of source profiles and this issue remains a large source of uncertainty in BC and OC inventory development.

Acknowledgements

This work was partially supported by the California Air Resources Board (ARB) Project No. 04-307 and the Strategic Environmental Research and Development Program (SERDP) Project WP-1336. The statements and conclusions in this report are those of the authors and not necessarily those of the ARB or the U.S. Department of Defense.

Appendix. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.atmosenv.2011.07.011.

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