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Particulate emission factors for mobile fossil fuel and biomass combustion sources

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ABSTRACT

PM emission factors (EFs) for gasoline- and diesel-fueled vehicles and biomass combustion were measured in several recent studies. In the Gas/Diesel Split Study (GD-Split), $PM_{2.5}$ EFs for heavy-duty diesel vehicles (HDDV) ranged from 0.2 to ~2 g/mile and increased with vehicle age. EFs for HDDV estimated with the U.S. EPA MOBILE 6.2 and California Air Resources Board (ARB) EMFAC2007 models correlated well with measured values. $PM_{2.5}$ EFs measured for gasoline vehicles were ~two orders of magnitude lower than those for HDDV and did not correlate with model estimates. In the Kansas City Study, $PM_{2.5}$ EFs for gasoline-powered vehicles (e.g., passenger cars and light trucks) were generally <0.03 g/mile and were higher in winter than summer. EMFAC2007 reported higher $PM_{2.5}$ EFs than MOBILE 6.2 during winter, but not during summer, and neither model captured the variability of the measured EFs. Total PM EFs for heavy-duty diesel military vehicles ranged from 0.18 ± 0.03 and 1.20 ± 0.12 g/kg fuel, corresponding to 0.3 and 2 g/mile, respectively. These values are comparable to those of on-road HDDV. EFs for biomass burning measured during the Fire Laboratory at Missoula Experiment (FLAME) were compared with EFs from the ARB Emission Estimation System (EES) model. The highest $PM_{2.5}$ EFs (76.8 ± 37.5 g/kg) were measured for wet (>50% moisture content) Ponderosa Pine needles. EFs were generally <20 g/kg when moisture content was <20%. The EES model agreed with measured EFs for fuels with low moisture content but underestimated measured EFs for fuel with moisture content >40%. Average EFs for dry chamise, rice straw, and dry grass were within a factor of three of values adopted by ARB in California's San Joaquin Valley (SJV). Discrepancies between measured and modeled emission factors suggest that there may be important uncertainties in current $PM_{2.5}$ emission inventories.

1. Introduction

Particulate matter (PM) emissions affect the Earth's climate (MacCracken, 2008a, 2008b), visibility (Chow et al., 2002; Watson, 2002), surface soiling (Sabbioni and Brimblecombe, 2003; Sabbioni et al., 2003), crop productivity (Grantz et al., 2003), and human health (Chow et al., 2006; Mauderly and Chow, 2008; Pope, III and Dockery, 2006).

Annual emission rates are compiled by states, provinces, and countries (CARB, 2009a; Environment Canada, 2008; EPD, 2008; U.S. EPA, 2008a) in a bottom-up approach to estimate primary $PM_{2.5}$ and PM_{10} (PM mass with aerodynamic diameters less than 2.5 and 10 µm, respectively), carbon monoxide (CO), reactive organic gasses (ROG, sometimes termed total non-methane hydrocarbons [NMHC] or volatile organic compounds [VOCs]), sulfur dioxide (SO_2), oxides of nitrogen (NO_x), and sometimes ammonia (NH_3). These inventories are usually expressed as tons/year or tonnes/year and are derived as the products of emission factors (EFs) and activities for different source categories (Mobley et al., 2005). $PM_{2.5}$ and PM_{10} mass emissions can be sub-divided into chemical components by applying source profiles (Watson, 1984; Watson et al., 2008a), or the mass fraction of each measured chemical component in primary emissions for each source category (CARB, 2009b; U.S. EPA, 2007).

The majority of PM mass from combustion sources such as engine exhaust and biomass burning is in the $PM_{2.5}$ fraction (Lighty et al., 2000; Lloyd and Cackette, 2001), and emission rates and compositions have changed as new fuels and combustion technologies have been adopted (Chow, 2001). In 2006, mobile fossil fuel and biomass combustion sources accounted for 16 and 47% of $PM_{2.5}$ emissions, respectively, and 43 and 53% of black carbon (BC) emissions,

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respectively, in California (Chow et al., 2010). $PM_{2.5}$ EFs were measured in the Gas/Diesel Split Study (GD-Split; Fujita et al., 2007a, 2007b), the Kansas City Study (Kishan et al., 2006; Nam et al., 2008; U.S. EPA, 2008b, 2008c), the Strategic Environmental Research and Development Program (SERDP; Watson et al., 2008b), and the Fire Laboratory at Missoula Experiment (FLAME; McMeeking et al., 2008). Measurements from these studies are summarized, evaluated, and compared with those from California's emission inventory. Because $PM_{2.5}$ from engine exhaust and biomass burning are primarily composed of organic and elemental carbon (OC and EC), accurate emission estimates are needed to evaluate future climate-related emission control strategies (Bond and Sun, 2005; Jacobson, 2002) as well as to attain National Ambient Air Quality Standards (Bachmann, 2007; Chow et al., 2007a) for $PM_{2.5}$.

2. Emission characterization studies

2.1. Diesel and gasoline engine emission factors

The GD-Split Study measured exhaust from 53 light-duty vehicles (52 gasoline- and 1 diesel-fueled) and 34 light-, medium-, and heavy-heavy-duty diesel-fueled vehicles (HDDV). Dynamometer emission tests were conducted at the Ralphs Grocery distribution center in Riverside, California, during the summer of 2001 (June 2–23 for light-duty gasoline- and diesel-fueled vehicles and from July 20 to September 19 for HDDV). Emissions were sampled into a constant-volume sampler with continuous monitoring for CO, CO_2, NMHC, and $NO_{x,}$ and integrated filter sampling for $PM_{2.5}$ mass, elements, ions, OC, EC, and organic compounds. $PM_{2.5}$ emission rates were estimated using the MOBILE 6.2 (Cook et al., 2007; U.S. EPA, 2008d) and EMFAC2007 (CARB, 2007) emission models under conditions corresponding to those in the GD-Split Study tests (Fujita et al., 2007a, 2007b). The EMFAC2007 model considers technology group and odometer mileage in addition to vehicle model year. The MOBILE 6.2 model accounts for vehicle type and age but omits the influence of fuel type, mileage, driving mode, and vehicle maintenance (Rakha et al., 2003; McCarthy et al., 2006). The MOtor Vehicle Emission Simulator (MOVES) model (http://www.epa.gov/otaq/models/moves/index.htm) improves on MOBILE 6.2, but was not available at the time of this analysis. MOBILE 6.2 calculates EFs in grams per vehicle mile traveled (g/VMT) for $PM_{2.5}$ mass, lead (Pb), sulfate ($SO_4^{=}$), OC, and EC from gasoline- and diesel-engine exhaust, as well as for brake and tire wear. EMFAC2007 estimates non-speciated $PM_{2.5}$ and PM_{10} EFs (in g/VMT). To facilitate comparisons of model estimates with dynamometer measurements, which only account for tailpipe emissions, MOBILE 6.2 $PM_{2.5}$ EFs were calculated from the sum of Pb, $SO_4^{=}$, OC, and EC emissions for gasoline- and diesel-fueled vehicles.

Gasoline-fueled vehicles were operated according to a modified California Unified Driving Cycle Schedule (UDC; DieselNet, 2008). The UDC is more aggressive in terms of acceleration and maximum speeds than the Federal Test Procedure (FTP), especially during the hot-stabilized portion of the cycle. The gasoline-fueled vehicles were tested under “Warm Start” (WS) and “Cold Start” (CS) cycles. The Hot-City-Suburban (HCS) Heavy Vehicle Route and Highway Cycle (HW) were used in HDDV tests. Additional HDDV test cycles included the Cold-City-Suburban (CCS) Heavy Vehicle Route, hot idle (ID) and cold idle (CID) periods, a City-Suburban Heavy Vehicle Route with Jacobs Brake (CSJ), and a Heavy-Duty Urban Dynamometer Driving Schedule (UDDS). Busses were tested on the HCS and Manhattan Cycle (MC) for Transit Busses cycles.

Observed and model-estimated $PM_{2.5}$ EFs for diesel-fueled vehicles are presented in Fig. 1. Because some EFs from the GD-Split Study represented composites of exhaust from more than one vehicle, the corresponding MOBILE 6.2 and EMFAC2007 model estimates are presented as ranges. Table 1 includes 23 gasoline-fueled vehicle

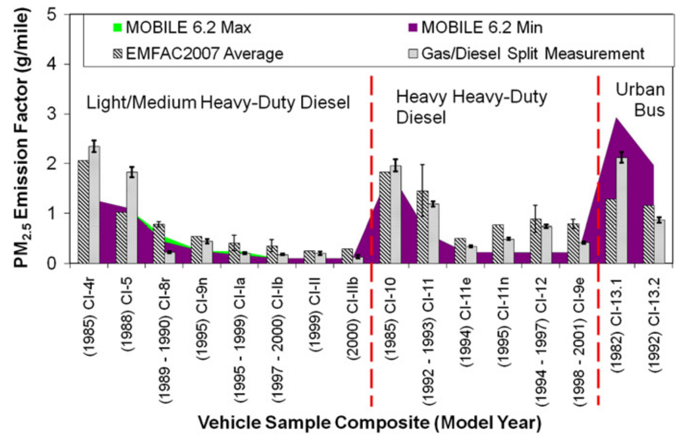

Fig. 1. Comparisons of measured diesel-fueled vehicle $PM_{2.5}$ emission factors (EFs) for the Hot City-Suburban route (HCS) driving cycle during the Gas/Diesel Split Study with MOBILE 6.2 and EMFAC 2007 model estimates for the Federal Test Procedure (FTP) cycle for each diesel group. See Table 1 for vehicle identification codes and composite information. Composites in each diesel group (light/medium heavy-duty, heavy heavy-duty, and urban bus) are ordered by the average vehicle model year. Error bars associated with the Gas/Diesel Split Study data indicate measurement uncertainties.

sample composites tested under WS and CS cycles, and 17 diesel-fueled vehicle sample composites tested under various cycles. Because the HCS cycle is common for all heavy-duty diesel vehicles in this study (Table 1), HCS $PM_{2.5}$ EFs are compared with MOBILE 6.2 and EMFAC2007 model estimates (for the FTP cycle) in Fig. 1 with the understanding that EFs for other cycles may differ (Fujita et al., 2007b).

Fig. 1 shows that the EMFAC2007 model slightly overestimated diesel-fueled vehicle emissions for GD-Split Study tests, especially for low emitters, but the overall agreement was good ($r^2 = 0.8$) considering the variability among individual vehicles. MOBILE 6.2 underestimated measured diesel vehicle EFs (within an order of magnitude), and correlation with measurements was moderate ($r^2 = 0.63$). Differences between minimum and maximum EF estimates by MOBILE 6.2 were small.

Both MOBILE 6.2 and EMFAC2007 estimated an increase in diesel vehicle $PM_{2.5}$ EFs with vehicle age (i.e., the difference between calendar year [2001] and vehicle model year), as shown in Fig. 2. Inter-cycle comparisons of measured EFs for typical EMFAC2007 medium heavy-duty vehicles (14,001–33,000 lbs) and heavy-heavy-duty vehicles (>33,000 lbs) are presented in Fig. 3. Both CCS and HCS cycles produced similar EFs which were about double those of HW cycle EFs. Sample composite CI-9e (Table 1) on the UDDS (i.e., FTP) cycle produced an EF ~20% lower than those measured on the HCS or CCS cycles.

The GD-Split Study gasoline-fueled vehicles were either passenger cars (LDA) or light-duty trucks (LDT). Their emissions were often mixed in a composite sample (Table 1). MOBILE 6.2 and EMFAC2007 reported distinct EFs for LDA and LDT vehicles, resulting in a wider range of EFs. Information on vehicle maintenance was not available and is not reflected in MOBILE 6.2 and EMFAC2007 EF estimates. The comparisons in Fig. 4 show that measured EFs for the WS and CS cycles were more variable than the modeled EFs, especially for vehicles manufactured before 1989 (Fig. 4a). A few high-emitting vehicles (often referred to as smokers) produced clear outliers (see footnote to Fig. 4), and all vehicles manufactured after 1995 displayed lower measured than modeled EFs.

Fig. 5 compares the GD-Split Study $PM_{2.5}$ EFs for gasoline-fueled vehicles under WS and CS cycles. EFs for CS (Fig. 5b) were higher than those for WS (Fig. 5a) with a few exceptions. Modeled and measured diesel-fueled vehicle EFs from Fig. 2 are superimposed in