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Speciated PM₁₀ Emission Inventory for Delhi, India

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ABSTRACT

Emission inventories can serve as a basis for air quality management programs. The focus has been mainly on building inventories for criteria pollutants including particulate matter (PM). Control efforts in developing countries are mostly limited to total suspended particles (TSP) and/or PM₁₀. Since the adverse effects of PM₁₀ depend on its chemical composition, it is important to control emissions of toxic species. The first step is to identify key pollution sources and estimate quantities of various chemical species in emissions. This paper presents a speciated PM₁₀ emission inventory for Delhi, the capital and one of the most polluted cities in India. An established PM₁₀ inventory for Delhi in conjunction with source profiles was used to estimate emissions of major PM₁₀ components including organic and elemental carbon (OC and EC, respectively), sulphates (SO₄²⁻), and nitrates (NO₃⁻), as well as selected toxic trace metals (i.e., Pb, Ni, V, As, and Hg), some of which are subject to India's National Ambient Air Quality Standards (NAAQS). For the base year of 2007, emission estimates for PM₁₀ mass, OC, EC, SO₄²⁻, and NO₃⁻ are 140, 22, 6.4, 2.8, and 2.1 tonnes/day (TPD; 1 tonne = 1000 kg), respectively. Emissions of Pb, Ni, V, As, and Hg are estimated to be 203, 43, 37, 26, and 9.4 kg/day, respectively. This inventory underestimated Pb and Hg emissions because sources of PM₁₀ from unorganized secondary lead smelters are not specifically identified and gas-to-particle conversion of Hg is not accounted for.

Keywords: Emission inventory; Delhi; Speciated PM₁₀.

INTRODUCTION

Emission inventories (EIs) provide a basis for understanding primary pollutant sources and for prioritizing pollution control measures (Mobley et al., 2005; Miller et al., 2006). While compiled over defined urban- to regionalscales to track long-term emission trends (U.S.EPA, 2010), annualized EIs can be apportioned in space and time to support air quality modeling that relates emissions to ambient concentrations (Hogrefe et al., 2003). Such modeling facilitates the selection of emission reduction strategies and evaluation of adverse effects on human health, visibility, climate, ecosystems, and cultural heritage (Watson, 2002; Pope, III and Dockery, 2006; MacCracken, 2008; Mauderly and Chow, 2008; Chow and Watson, 2011; Hu et al., 2011). Most EIs are developed for criteria pollutants such as carbon monoxide (CO), sulphur dioxide (SO₂), oxides of nitrogen (NO_x), total suspended particles (TSP) and/or PM₁₀ and

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PM_{2.5} (particles with aerodynamic diameters, d_{p} , < 10 and 2.5 micrometers [µm], respectively). Precursors to the formation of ozone (O₃) and secondary PM, such as volatile organic compounds (VOCs) and ammonia (NH₃), are sometimes included in the EIs. Long-lived greenhouse gases and short-lived climate forcers such as black and brown carbon are also being compiled on regional, continental, and global scales (Bond *et al.*, 2004; Junker and Liousse, 2008; Chow *et al.*, 2010; Lamarque *et al.*, 2010; Chow *et al.*, 2011; Watson *et al.*, 2011; Wang *et al.*, 2012).

Most inventory efforts in India (Gargava and Aggarwal, 1999; Garg *et al.*, 2002; Reddy and Venkataraman, 2002a, b; Gurjar *et al.*, 2004; Dalvi *et al.*, 2006; Mohan *et al.*, 2007; Gurjar *et al.*, 2008; Behera *et al.*, 2011; Lu *et al.*, 2011; Sahu *et al.*, 2011) have focused on primary pollutants related to India's National Ambient Air Quality Standards (NAAQS) (Central Pollution Control Board [CPCB], 2009). The Indian NAAQS include: 1) annual average PM₁₀ of 60 μ g/m³ and PM_{2.5} of 40 μ g/m³; 2) 24-hour average PM₁₀ of 100 μ g/m³ and PM_{2.5} of 60 μ g/m³, 98th percentile for one year, with no exceedance on two consecutive days; 3) annual average PM lead (Pb) of 0.5 μ g/m³, and benzo(a)pyrene (BaP) of 0.001 μ g/m³; and 4) 24-hour average PM Pb of

 $1.0 \ \mu\text{g/m}^3$, 98^{th} percentile for one year, with no exceedance on two consecutive days (CPCB, 2009).

Speciated PM and VOC EIs are needed to estimate O₃ and secondary aerosol formation from primary emissions, evaluate exposure to toxic pollutants, estimate adverse environmental effects, and compare emission estimates with ambient concentrations. Bhanarkar et al. (2005) reported 2001-2002 industrial PM emissions for toxic metals in Greater Mumbai, India, finding that vanadium (V) constituted ~57% of all toxic metals in PM, mostly derived from residual oil combustion. More up-to-date speciated inventories that address additional sources and chemical species are needed for India's major population centers. This study derives a speciated PM₁₀ inventory for primary species, including organic and elemental carbon (OC and EC, respectively), sulphates (SO_4^{2-}) , nitrates (NO_3^{-}) , and metals (i.e., Pb, Ni, V, As, and mercury [Hg]) in the Delhi metropolitan area for 2007.

STUDY AREA

Delhi, the Capital and economic center of India, is one of the world's most polluted megacities (Chow *et al.*, 2004; Molina and Molina, 2004). Despite remediation actions including implementation of the world's largest compressed natural gas (CNG)-fueled public transportation system, air pollution remains a critical issue (Goyal and Sidhartha, 2003; Ravindra *et al.*, 2006; Chelani and Devotta, 2007; Khillare *et al.*, 2008; Saxena and Ghosh, 2010). Measurements from 2001 to 2010 indicate increasing PM₁₀ trends with annual averages at nine sites almost four times the NAAQS of 60 μ g/m³ Annual average PM_{2.5} concentrations ranged from 72 to 107 μ g/m³ at six sites in 2010, far exceeding the 40 μ g/m³ NAAQS (CPCB, 2009).

The Delhi emissions domain (i.e., latitudes 28°24' 17" and 28°53'00 N and longitudes 76°50'24" and 77°20'37" E), encompasses an area of approximately 1500 km^2 . Its population increased from ~13.8 million in 2001 to ~17 million in 2010 (Census of India, 2012); the number of motor vehicles rose from ~3.6 million in 2002, ~5.2 million in 2007, and to ~6.5 million in 2011. The vehicle fleet consists of 63.6% two-wheelers and 30.7% gasoline-fueled passenger vehicles, as well as minor amounts of light commercial vehicles (1.6%), three-wheelers (1.4%), trucks (1.1%), buses (0.8%), and diesel-fueled vehicles (0.5%)(NEERI, 2010). Energy demand increased from ~3100 megawatts (MW) in 2007 to ~5900 MW in 2012 (National Capital Region [NCR] Planning Board. 2001). This growth has the potential to offset many of the emission reduction measures applied to individual sources.

METHODS

A study was conducted in Delhi during 2007 to: 1) characterize PM_{10} mass and its chemical constituents at 10 monitoring sites, 2) develop micro- and city-wide PM_{10} emission inventories, and 3) conduct dispersion and receptor modeling to estimate the source contribution to PM_{10} and other criteria pollutants (NEERI, 2010). Samples were

collected in summer (May–June), post-monsoon (October– November), and winter (December–January) periods on 20 days during each season. PM_{10} samples were analyzed for mass, carbon (i.e., OC and EC), ions (i.e., SO_4^{2-} , and NO_3^{-}), and trace metal (i.e., Pb, Ni, V, As, and Hg) concentrations.

Emission inventories were constructed in the NEERI (2010) study for PM₁₀ mass and other criteria pollutants in 2×2 km² zones of influence (Chow *et al.*, 2002) surrounding each monitoring site. These "micro-inventories" were input to the U.S. EPA Industrial Source Complex-Short Term (ISCST3) dispersion model (U.S. EPA, 1995a, b) to estimate emissions and to compare with measured ambient concentrations. Monitoring sites were located in representative neighborhoods so that area-source activities could be extrapolated to similar neighborhoods throughout the metropolis. These monitoring sites represented various land-use (e.g., industrial, residential, and commercial areas) and activity profiles (e.g., traffic). PM₁₀ emission rates were estimated by multiplying emission factors for different source types by their corresponding activities. Table 1 summarizes 30 stationary, area, and vehicular source categories that were included in the 2007 emission inventory.

The methodologies used to estimate emission factors and activity levels for the various sources are described in NEERI (2010). Identification of stationary and area sources, fuel use, and population were collected from surveys of government and non-government departments and agencies and from direct observation, as in the case of open burning. For motor vehicles, traffic volumes were counted for different categories of roads (i.e., arterial, main, and feeder roads). Vehicle kilometers traveled (VKT) were estimated by multiplying traffic volume (number of vehicles) by the effective road length for various road categories. Data on fuel use and vehicle age were assembled through surveys at parking lots and fueling stations (NEERI, 2010). Delhi contains many gasoline-fueled two- and CNG-fueled threewheeled vehicles that emit carbon in the ultrafine $(d_p < 0.1)$ μ m) and fine (d_p < 2.5 μ m) particle size range (Apte *et al.*, 2011). These representative activity data were associated with city land use and roadway plans. Re-suspended dust from paved roads was estimated from silt loading, average weight of vehicles (tonnes/day), road length, and rainfall frequency. Average silt loading has been estimated to be in the range of 0.21 to 7.0 g/m^2 depending on the road condition. Re-suspended road dust was estimated for each of the road categories.

A city-wide inventory for stationary, area, and vehicular sources was developed incorporating the 30 fuel usage source categories identified in Table 1, population, and activity levels. Vehicle emissions were extrapolated from the micro-inventories at the 10 monitoring sites by re-scaling road lengths in each road category and estimating emissions per unit length of road for the entire city. Emission factors for stationary and area sources were derived from U.S. AP-42 (U.S. EPA, 2006) and Reddy and Venkataraman (2002a, b). Emission factors for motor vehicles were taken from the Automotive Research Association of India (ARAI; 2007) and CPCB (2011). These emission factors are presented in Appendix 3 of NEERI (2010).

Source No.	Source ID	Source Description
Stationary and	Area Sources	
1	PP_Coal	Coal-fired power plant
2	PP_NG	Natural Gas-fired power plant
3	Ind_NG	Natural Gas-fueled industrial boiler
4	Ind_FO	Fuel Oil (Diesel)-fueled industrial boiler
5	LPG_Dom	Domestic cooking with LPG ^a
6	Kero_Dom	Domestic cooking with kerosene
7	Wood_Dom	Domestic cooking with wood
8	Coal_Dom	Domestic cooking with coal
9	LPG_Hotel ^b	Commercial cooking with LPG
10	Kero_Hotel ^b	Commercial cooking with kerosene
11	Wood_Hotel ^b	Commercial cooking with wood
12	Coal_Hotel ^b	Commercial cooking with coal
13	Wood_Bak ^b	Commercial bakeries with wood
14	Crmt_wood	Crematoria with wood
15	Waste_Inc	Waste incinerator
16	SW_burning	Open solid waste (e.g., refuse and biomass) burning
17	Construction	Construction activities
18	Genset_K	Generator set using kerosene-fueled engine
19	Genset_D	Generator set using diesel-fueled engine
20	PvdRd	Paved road dust
Vehicular Sour	ces	
21	2W-2S	Two wheeler (2W) with gasoline-fueled two stroke (2S) engine
22	2W-4S	Two wheeler with gasoline fueled four stroke (4S) engine
23	2W	Two wheeler including both 2W-2S and 2W-4S
24	3W_CNG	Compressed natural gas (CNG)-fueled three wheeler
25	LCV	Diesel-fueled light commercial vehicles
26	4W_P	Gasoline-fueled passenger vehicles
27	4W_D	Diesel-fueled passenger vehicles
28	Truck_D	Diesel-fueled trucks
29	Bus D	Diesel-fueled bus

CNG-fueled bus

Table 1. Source categories in the Delhi PM₁₀ emission inventory (NEERI, 2010).

^a Liquefied petroleum gas.

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^b Hotels, restaurants, and small dining outlets.

Bus CNG

Chemically-speciated PM_{10} emission rates were estimated by multiplying PM_{10} mass emission rates from the specific source sub-categories in Table 1 by the corresponding source profiles. Source profiles are the fractional abundances of chemical species with respect to emitted PM_{10} mass (Reff *et al.*, 2009; Chow *et al.*, 2010; 2011; Watson *et al.*, 2012). PM_{10} source profiles specific to India, presented in Table 2, were measured by Sethi and Patil (2008a, b) for stationary and area sources, and by ARAI (2009) for vehicular sources. Emission rates for chemical species were calculated using the following equation:

$$\mathbf{E}_{i,j} = \mathbf{P}\mathbf{M}_j \times \mathbf{F}_{i,j},\tag{1}$$

where $E_{i,j}$ is the emission rate in kg/day of species i from source j; PM_j is the emission rate in kg/day of PM_{10} from source j; and $F_{i,j}$ is the mass fraction of species i from source j.

Table 3 summarizes the chemically-speciated PM_{10} emission inventory. Apart from power plants, industrial boilers (Source Nos. 3 and 4) are classified as small-scale

industries (SSI; NEERI, 2010), which account for ~1.6% of the total PM₁₀ emissions in Delhi. These industries use mostly fuel oil (diesel) and natural gas. In the absence of specific data on fuel usage, 80 and 20% of SSI combustion was apportioned to fuel oil and natural gas, respectively. For the generator sets (Source Nos. 18 and 19) used during sporadic power outages, emissions were equally distributed between diesel- and kerosene-fueled engines. In December 2007, one coal-fired power plant (Indraprastha Power Station) was permanently shut down. Emissions (~5 tonnes per day [TPD]; 1 tonne = 1000 kg) from this power plant and from a few smaller sources (e.g., locomotives, dung cakes, etc.) were excluded due to non-availability of indigenous source profiles. However, 95% of the PM₁₀ emission load was accounted for and used to determine the speciated emission inventory.

To better understand the sectoral emission distributions, the 30 source sub-categories in Table 3 are grouped into 12 primary source categories based on their similarities in source type and/or fuel usage. These include: power plants; small-scale industries; domestic cooking and heating (i.e.,

	L	Table 2. Source	profiles (perce	nt of PM ₁₀ ma	ss) and associa	ted uncertainties	corresponding .	to the source catego	ories in Table 1.	
Source No. ^a	Source ID	OC	EC	SO_4^{2-}	NO_3^-	Pb	Ni	Λ	As	Hg
1	PP_Coal^a	8.71 ± 0.44	1.88 ± 0.94	1.43 ± 0.12	6.87 ± 0.51	0.100 ± 0.040	0.090 ± 0.037	0.0523 ± 0.0117	0.0257 ± 0.0185	0.0070 ± 0.0050
0	PP_NG	26.54 ± 1.33	7.48 ± 0.90	6.01 ± 0.52	39.96 ± 2.97	0.114 ± 0.170	0.304 ± 0.185	0.1273 ± 0.0533	0.4732 ± 0.1167	0.0369 ± 0.0265
ς	Ind_NG	26.54 ± 1.33	7.48 ± 0.90	6.01 ± 0.52	39.96 ± 2.97	0.114 ± 0.170	0.303 ± 0.185	0.1273 ± 0.0533	0.4732 ± 0.1167	0.0369 ± 0.0265
4	Ind FO	3.73 ± 0.19	16.42 ± 0.82	30.06 ± 2.13	0.04 ± 0.01	0.010 ± 0.007	0.299 ± 0.027	0.6553 ± 0.0477	0.0241 ± 0.0052	0.0015 ± 0.0011
5	LPG Dom	23.21 ± 1.16	2.82 ± 0.14	7.74 ± 0.63	0.25 ± 0.18	0.446 ± 0.139	0.056 ± 0.121	0.0488 ± 0.0350	0.3278 ± 0.0814	0.0258 ± 0.0185
9	Kero_Dom	96.90 ± 4.85	52.68 ± 2.63	12.41 ± 0.98	0.32 ± 0.23	0.237 ± 0.210	0.282 ± 0.217	0.3148 ± 0.0743	0.2039 ± 0.1196	0.0444 ± 0.0319
٢	Wood Dom	48.45 ± 2.42	8.00 ± 0.40	0.83 ± 0.07	0.24 ± 0.05	0.471 ± 0.062	0.022 ± 0.034	0.0053 ± 0.0093	0.1054 ± 0.0234	0.0072 ± 0.0051
8	Coal_Dom	51.72 ± 2.59	7.81 ± 0.39	5.93 ± 0.44	0.29 ± 0.06	1.572 ± 0.136	0.022 ± 0.034	0.0137 ± 0.0099	0.0267 ± 0.0192	0.0073 ± 0.0052
6	LPG Hotel	23.21 ± 1.16	2.82 ± 0.14	7.74 ± 0.63	0.25 ± 0.18	0.446 ± 0.139	0.056 ± 0.121	0.0488 ± 0.0350	0.3278 ± 0.0814	0.0258 ± 0.0185
10	Kero_Hotel	96.90 ± 4.85	52.68 ± 2.63	12.41 ± 0.98	0.32 ± 0.23	0.237 ± 0.210	0.282 ± 0.217	0.3148 ± 0.0743	0.2039 ± 0.1196	0.0444 ± 0.0319
11	Wood Hotel	48.45 ± 2.42	8.00 ± 0.40	0.83 ± 0.07	0.24 ± 0.05	0.471 ± 0.062	0.022 ± 0.034	0.0053 ± 0.0093	0.1054 ± 0.0234	0.0072 ± 0.0051
12	Coal Hotel	51.72 ± 2.59	7.81 ± 0.39	5.93 ± 0.44	0.29 ± 0.06	1.572 ± 0.136	0.022 ± 0.034	0.0137 ± 0.0099	0.0267 ± 0.0192	0.0073 ± 0.0052
13	Wood Bak	48.45 ± 2.42	8.00 ± 0.40	0.83 ± 0.07	0.24 ± 0.05	0.471 ± 0.062	0.022 ± 0.034	0.0053 ± 0.0093	0.1054 ± 0.0234	0.0072 ± 0.0051
14	Crmt wood	48.45 ± 2.42	8.00 ± 0.40	0.83 ± 0.07	0.24 ± 0.05	0.471 ± 0.062	0.022 ± 0.034	0.0053 ± 0.0093	0.1054 ± 0.0234	0.0072 ± 0.0051
15	Waste Inc	76.04 ± 3.80	1.84 ± 0.09	0.37 ± 0.04	3.70 ± 0.28	2.946 ± 0.217	0.014 ± 0.013	0.0718 ± 0.0079	0.0101 ± 0.0073	0.0028 ± 0.0020
16	SW_burning	52.06 ± 2.25	1.44 ± 0.06	0.25 ± 0.02	0.01 ± 0.01	0.017 ± 0.006	0.001 ± 0.005	0.0020 ± 0.0014	0.0038 ± 0.0027	0.0010 ± 0.0007
17	Construction	8.79 ± 0.44	1.42 ± 0.07	3.21 ± 0.23	0.80 ± 0.06	0.043 ± 0.043	0.007 ± 0.007	0.0030 ± 0.0030	0.0059 ± 0.0059	0.0016 ± 0.0016
18	Genset_K	63.40 ± 3.17	0.74 ± 0.04	0.20 ± 0.02	0.28 ± 0.03	0.076 ± 0.017	0.008 ± 0.013	0.0052 ± 0.0037	0.0084 ± 0.0072	0.0028 ± 0.0020
19	Genset_D	16.49 ± 0.82	32.54 ± 1.63	2.65 ± 0.27	8.45 ± 0.73	0.135 ± 0.148	0.097 ± 0.149	0.4749 ± 0.0682	0.1475 ± 0.0852	0.0316 ± 0.0227
20	PvdRd	9.18 ± 0.46	2.81 ± 0.14	1.32 ± 0.10	0.01 ± 0.01	0.135 ± 0.009	0.013 ± 0.004	0.0016 ± 0.0011	0.0032 ± 0.0022	0.0080 ± 0.0009
21	2W-2S	57.34 ± 4.64	3.10 ± 0.26	4.24 ± 0.51	0.86 ± 0.15	0.037 ± 0.010	0.000 ± 0.004	0.1919 ± 0.1919	0.0000 ± 0.0022	NA^{b}
22	2W-4S	43.22 ± 3.50	3.36 ± 0.28	2.67 ± 0.39	1.15 ± 0.13	0.105 ± 0.011	0.000 ± 0.005	0.8703 ± 0.0029	0.0000 ± 0.0025	NA
23	2W	50.28 ± 4.11	3.23 ± 0.27	3.45 ± 0.45	1.00 ± 0.14	0.071 ± 0.011	0.000 ± 0.000	0.5311 ± 0.0027	0.0000 ± 0.0024	NA
24	3W_CNG	43.73 ± 3.54	3.89 ± 0.32	2.14 ± 1.03	0.09 ± 0.35	0.013 ± 0.031	0.000 ± 0.013	0.0572 ± 0.0079	0.0000 ± 0.0069	NA
25	LCV	48.09 ± 3.90	23.95 ± 1.99	0.93 ± 0.07	0.24 ± 0.02	0.005 ± 0.001	0.001 ± 0.000	0.0123 ± 0.0003	0.0010 ± 0.0003	NA
26	$4W_P$	49.61 ± 4.02	12.48 ± 1.04	5.26 ± 0.58	1.55 ± 0.21	0.053 ± 0.020	0.020 ± 0.008	0.4495 ± 0.0051	0.0165 ± 0.0044	NA
27	$4W_D$	53.40 ± 4.33	18.49 ± 1.53	0.82 ± 0.06	0.16 ± 0.02	0.013 ± 0.001	0.001 ± 0.000	0.0001 ± 0.0003	0.0020 ± 0.0020	NA
28	Truck_D	50.42 ± 4.08	26.88 ± 2.23	2.91 ± 0.19	0.29 ± 0.03	0.011 ± 0.002	0.001 ± 0.001	0.0000 ± 0.0006	0.0005 ± 0.0005	NA
29	Bus_D	50.42 ± 4.08	26.88 ± 2.23	2.91 ± 0.19	0.29 ± 0.03	0.011 ± 0.002	0.001 ± 0.001	0.0000 ± 0.0006	0.0005 ± 0.0005	NA
30	Bus_CNG	29.57 ± 1.82	20.17 ± 1.24	8.33 ± 0.47	2.99 ± 0.19	0.025 ± 0.010	0.024 ± 0.004	0.0000 ± 0.0024	0.0000 ± 0.0021	NA
^a Source	Numbers 1 to	20 from Sethi	and Patil (2008	(); Source Num	bers 21 to 30 f	from ARAI (200	9) and CPCB (2	011).		
^b Inform	ation not avail	lable.	*					×		

Source No. ^a	Source_ID	PM ₁₀	OC	EC	TC	SO ₄ ²⁻	NO ₃ ⁻	Pb	Ni	V	As	Hg
I. Stationary	[,] and Area Sources ^b)										
Power Plant	Sources ^b											
1	PP Coal	25,290	2201.5	474.7	2676.2	362.7	1737.9	25.3	22.7	13.2	6.5	1.8
2	PP_NG	6	1.6	0.4	2.0	0.4	2.4	0.0	0.0	0.0	0.0	0.0
Sum of	Power Plant ^c	25,296	2203.1	475.2	2678.3	363.1	1740.3	25.3	22.8	13.2	6.5	1.8
Industrial Bo	oiler Sources											
3	Ind NG	447	118.5	33.4	151.9	26.8	178.5	0.4	0.4	0.2	0.0	0.0
4	Ind FO	1786	66.7	293.4	360.1	537.1	0.7	0.2	5.3	11.7	0.4	0.0
Sum of In	dustrial Boilers ^c	2233	185.2	326.8	512.0	563.9	179.2	0.6	5.7	11.9	0.4	0.1
Domestic Cooking Sources ^b												
5	LPG Dom	660	153.2	18.6	171.8	51.1	1.7	2.9	0.4	0.3	2.2	0.2
6	Kero Dom	100	96.9	52.7	149.6	12.4	0.3	0.2	0.3	0.3	0.2	0.0
7	Wood Dom	10,610	5140.0	848.7	5988.7	88.0	25.7	50.0	2.4	0.6	11.2	0.8
8	Coal Dom	230	119.0	18.0	136.9	13.6	0.7	3.6	0.1	0.0	0.1	0.0
Sum of Do	omestic Cooking ^c	11,600	5509.1	937.9	6447.0	165.1	28.4	56.8	3.1	1.2	13.6	1.0
Commercial												
9	LPG Hotel	130	30.2	3.7	33.8	10.1	0.3	0.6	0.1	0.1	0.4	0.0
10	Kero_Hotel	1	1.4	0.7	2.1	0.2	0.0	0.0	0.0	0.0	0.0	0.0
11	Wood_Hotel	20	9.7	1.6	11.3	0.2	0.0	0.1	0.0	0.0	0.0	0.0
12	Coal_Hotel	0	0.1	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Sum of Commercial Cooking ^c		152	41.4	6.0	47.4	10.4	0.4	0.7	0.1	0.1	0.5	0.0
Individual Sources ^b												
13	Wood_Bak ^c	120	58.1	9.6	67.7	1.0	0.3	0.6	0.0	0.0	0.1	0.0
14	Crmt_wood ^c	1300	629.8	104.0	733.8	10.8	3.2	6.1	0.3	0.1	1.4	0.1
15	Waste_Inc ^c	17	12.9	0.3	13.2	0.1	0.6	0.5	0.0	0.0	0.0	0.0
16	SW_burning ^c	1	0.7	0.0	0.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0
17	Construction ^c	12,290	1080.7	174.9	1255.6	394.0	98.9	5.3	0.9	0.4	0.7	0.2
Generator Se	ources ^b											
18	Genset_K	267	169.3	2.0	171.3	0.5	0.7	0.2	0.0	0.0	0.0	0.0
19	Genset_D	267	44.0	86.9	130.9	7.1	22.6	0.4	0.3	1.3	0.4	0.1
Sum o	f Generators ^c	534	213.3	88.9	302.2	7.6	23.3	0.6	0.3	1.3	0.4	0.1
Paved Road	Source ^b											
20	PvdRd ^c	77,275	7094.5	2173.1	9267.6	1016.8	6.9	104.6	9.9	1.2	2.5	6.1
II. Vehicula	r Sources ^b											
21	2W-2S	427	244.9	13.3	258.1	18.1	3.7	0.2	0.0	0.8	0.0	0.0
22	2W-4S	650	280.9	21.8	302.8	17.4	7.5	0.7	0.0	5.7	0.0	0.0
23	2W	1077	541.5	34.8	576.4	37.2	10.8	0.8	0.0	5.7	0.0	0.0
24	3W_CNG	582	254.5	22.6	277.2	12.5	0.5	0.1	0.0	0.3	0.0	0.0
25	LCV	2677	1287.3	641.3	1928.6	24.9	6.4	0.1	0.0	0.3	0.0	0.0
26	4W_P	165	81.9	20.6	102.4	8.7	2.6	0.1	0.0	0.7	0.0	0.0

Table 3. Speciated PM_{10} emission rates (kg/day) for Delhi in 2007.

^a See Table 1 for source category descriptions.

4W D

Bus D

Sum of Vehicles^c

Total

Truck_D

Bus^{CNG}

27

28

29

30

^b Italicized headers under Stationary and Area Sources indicate source sub-categories.

77.4

2240.1

303.0

3.0

145

4443

601

10

9700

^c Bold indicates the 12 primary source categories including sub-category summation and individual source categories.

26.8

1194.3

161.6

2.0

140,518^d 21,817.7 6400.7 28,218.4 2765.1

4788.8 2104.0

104.2

3434.5

464.6

5.0

6892.8

1.2

129.5

17.5

0.8

232.3

0.2

13.0

1.8

0.3

35.6

0.0

0.5

0.1

0.0

1.7

2117.0 202.6

0.0

0.0

0.0

0.0

0.1

43.2

0.0

0.0

0.0

0.0

7.1

36.6

0.0

0.0

0.0

0.0

0.1

26.2

0.0

0.0

0.0

0.0

0.0

9.4

^d The difference between this (140 TPD) and the 147 TPD listed in Table E.1 of http://www.cpcb.nic.in/Delhi.pdf is due to the closure of the Indraprastha Power Plant in December 2007 and the exclusion of a few smaller sources (e.g., locomotive, dung cake, etc.).

liquified petroleum gas [LPG], kerosene, wood, and coal); commercial cooking; wood burning; crematoria (wood); waste incineration; open solid waste burning; construction activities (i.e., building and road construction); generators (i.e., diesel- and kerosene-fueled); paved road dust; and motor vehicles (i.e., two- to four-stroke engines for twowheelers, gasoline- and diesel-fueled passenger vehicles, commercial vehicles, and diesel- and CNG-fueled buses).

RESULTS AND DISCUSSION

Emissions

As shown in Table 3, out of 140 TPD of PM_{10} emissions, carbon emissions (OC = 21.8 TPD, EC = 6.4 TPD) accounted for 20% of total PM_{10} , followed by primary (emitted directly as particles) SO_4^{2-} (2.8 TPD) and NO_3^- (2.1 TPD). Toxic metals accounted for a small fraction, which was dominated by Pb (203 kg/day), followed by Ni (43 kg/day), V (37 kg/day), As (26 kg/day), and Hg (9.4 kg/day).

Source attributions to daily PM₁₀ mass and chemical species among the 12 primary source categories are presented in Table 4. Paved road dust was the principal source of PM₁₀ (55%), OC (33%), EC (34%), SO₄²⁻ (37%), Pb (52%) and Hg (65%). Power plants, including both coal- and natural gas-fired, were the largest source of primary NO₃⁻ (82%), Ni (53%), and V (36%), the second largest contributors to Hg (19%) and As (25%), and the third largest source of Pb (13%). Industrial boilers, mainly burning heavy-duty diesel fuel oil, were the second largest source of SO_4^{2-} (20%) and V (33%), and the third largest source of Ni (13%). Lead was phased out as a gasoline additive starting in 1992 (CPCB, 2010) and vehicles were a minor source of Pb emissions (< 1%). Paved road dust was the major contributor to Hg (65%) and Pb (52%) only because it was the major contributor to PM_{10} (55%). This is also the case for OC, EC, and SO_4^{2-} . Table 2 shows that Pb only accounts for 0.135% of PM₁₀ in the paved road dust profile; two orders of magnitude lower than the Pb abundance (17.5%) found in secondary Pb smelter emissions (ARAI, 2009). The NEERI (2010) PM₁₀ inventory does not specifically identify emissions from unregistered secondary Pb smelting or reprocessing of lead-acid batteries, a known source of Pb emissions in Delhi (Gupt, 2012).

Domestic cooking, mainly using wood fuel, was the

largest emitter of As (52%) and the second largest emitter of Pb (28%), with abundant Hg (11%) emissions. Vehicle exhaust was also a major emitter of OC (22%), EC (33%), V (20%), and to a lesser extent, SO_4^{2-} (8%) and PM_{10} (7%). Aside from the missing sources in the inventory (Table 1) that inflate the percent species contribution (Table 4), uncertainties in the source sampling and chemical analyses also contribute to the deviations among distributions.

The ratio of ambient concentrations to source emissions (R) may be interpreted as a first-order comparative index to verify how well the inventory accounts for chemical species concentration (Fujita *et al.*, 1992; Brown, 2010). For primary sources, an R greater than unity suggests underestimation of source emissions in the inventory. Table 5 compares the annual average PM₁₀ and chemical concentrations at the 10 monitoring sites with daily source emission rates in 2007. India's annual NAAQS levels are also listed for comparison. The network-wide annual average PM₁₀ mass and chemical species concentrations exceeded the NAAQS by three- to twelve-fold.

Table 5 shows R = 2.9 for PM_{10} mass, suggesting underestimation of emission rates in the inventory, which might be partially due to the uninventoried wind-blown dust emissions. By contrast, R for OC and EC are 1.0 and 0.6, respectively. High ratios for $SO_4^{2-}(9.8)$ and $NO_3^{-}(8.5)$ are expected, since these species are mostly secondary aerosol and may be formed during long-range pollution transport. R for trace elements is high, in the range of 2 to 9. In addition to missing sources for Pb (R = 9.2), the inventory is mainly based on fuel use rather than industrial processes, which can cause large uncertainties for trace element emission estimates. This can also explain moderately high ratios for Ni (2.1), V (1.9), and As (2.7). The elevated R for Hg (8.6) is anomalous. Mercury is present mainly as gaseous elemental Hg, while reactive gaseous Hg in coal combustion and other industrial emissions can enter the particle phase through atmospheric oxidation processes (Galbreath et al., 1996; Amos et al., 2012). Gas-to-particle

Table 4. Source contributions to daily PM₁₀ mass and chemical species in Delhi, India.

Primary Source	Source		Perc	ent of To	otal PM ₁₀	Mass an	d Chemi	cal Spec	ies Emiss	sions	
Category ^a	No. ^b	PM ₁₀	OC	EC	SO_4^{2-}	NO_3^-	Pb	Ni	V	As	Hg
Power Plants	1–2	18.0	10.1	7.4	13.1	82.2	12.5	52.7	36.2	24.91	18.9
Industrial Boilers	3–4	1.6	0.8	5.1	20.4	8.5	0.3	13.3	32.7	1.64	0.6
Domestic Cooking	5-8	8.3	25.3	14.7	6.0	1.3	28.0	7.1	3.4	51.89	10.6
Commercial Cooking	9-12	0.1	0.2	0.1	0.4	0.0	0.3	0.2	0.2	1.72	0.4
Wood_Bak	13	0.1	0.3	0.1	0.0	0.0	0.3	0.1	0.0	0.48	0.1
Crmt_wood	14	0.9	2.9	1.6	0.4	0.1	3.0	0.7	0.2	5.22	1.0
Waste_Inc	15	0.0	0.1	0.0	0.0	0.0	0.2	0.0	0.0	0.01	0.0
SW_burning	16	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.00	0.0
Construction	17	8.7	5.0	2.7	14.2	4.7	2.6	2.1	1.0	2.75	2.1
Generators	18-19	0.4	1.0	1.4	0.3	1.1	0.3	0.7	3.5	1.51	1.0
PvdRd	20	55.0	32.5	34.0	36.8	0.3	51.6	22.9	3.3	9.57	65.4
Vehicles	21 - 30	6.9	21.9	32.9	8.4	1.7	0.8	0.3	19.5	0.31	0.0

^a See Table 3 for the 12 primary source categories. Emission rates from source sub-category summations and individual source categories were normalized to the total emissions for PM_{10} and chemical speciation to derive percent source contribution.

^b See Table 1 for Source Number and description.

	India Annual	Annual Average	Daily Source	Ratio of Ambient
Observable	Average NAAQS ^a	Ambient Concentration ^b	Emissions	Concentration/Source Emissions
	$(\mu g/m^3)$	$(\mu g/m^3)$	(tonnes/day [TPD])	(R)
PM_{10}	60	412	140	2.9
OC	NA ^c	22.6	21.8	1.0
EC	NA	4.0	6.4	0.6
$\mathrm{SO_4}^{2-}$	NA	27.4	2.8	9.8
NO_3^-	NA	17.8	2.1	8.5
Pb	0.5	1.84	0.20	9.2
Ni	0.02	0.091	0.043	2.1
V	NA	0.071	0.037	1.9
As	0.006	0.071	0.026	2.7
Hg	NA	0.081	0.0094	8.6

Table 5. Comparison between annual average ambient concentrations for the 10 monitoring sites and daily source emission rates for Delhi in 2007.

^a National Ambient Air Quality Standards.

^b Averages over three seasons, 20 days per season at the 10 PM₁₀ monitoring sites. Seasons are defined as: summer (May–June), post-monsoon (October–November), and winter (December–January).

^c Information not available.

transformation of Hg is not accounted for in the PM_{10} inventory.

Emission rates for vehicular sources in Table 3 were estimated for vehicle types/fuel types (e.g., diesel-fueled buses, trucks, and vehicles; CNG-fueled three-wheelers and buses; and gasoline-fueled two-wheelers and vehicles). Fig. 1 presents the contributions of diesel-, CNG-, and gasoline-fueled vehicles to PM₁₀ mass and chemical species within the vehicular source category. Diesel-fueled vehicles contributed nearly all of the vehicular EC (96%) and most of the PM_{10} mass (81%), OC (82%), primary SO_4^2 (75%), and Ni (71%). Heavy-duty diesel vehicles (i.e., mostly trucks, Source No. 28 in Table 3) were the largest emitters among on-road vehicles, contributing 47% of OC, 57% of EC, 56% of primary SO_4^{2-} , 37% of primary NO_3^{-} , 30% of Pb, and 35% of Ni emissions. Light commercial diesel vehicles (LCV, Source No. 25) contributed in the range of 10-30% of vehicular emissions.

Fig. 1 shows that gasoline-fueled vehicles contributed nearly all of the V (92%) and the majority of the Pb (54%). Within the vehicular source category, gasoline-fueled two-wheelers (about 65% of total vehicles) contributed ~80% to V and 48% to Pb emissions. None of the vehicular source profiles contain Hg (Table 2), thus, this source category does not contribute to Hg. The large contribution of two-wheeled four-stroke vehicles (2W-4S, Source No. 22) to V (~80%) seems incongruous. PM₁₀ V is typically enriched in emissions from residual or heavy-duty fuel oil (Olmez and Gordon, 1985). Yet, Table 2 indicates that the V content in the 2W-4S source profile is 0.87% of PM₁₀, the highest in any of the available source profiles for India.

Table 6 presents PM_{10} source attributions for seven emission sources by fuel types (i.e., including vehicular categories but excluding road dust, construction, and nonfuel specific sources [i.e., open burning and waste incineration]) in the city-wide inventory. PM_{10} mass emissions originated mainly from coal (53%) and wood (25%) combustion; OC from wood (43%) and diesel-fuel (29%) combustion; and EC from diesel fuel (56%) and wood (26%) combustion. While wood and coal combustion account for 62 and 31%, respectively, of Pb in the inventory, coal combustion dominates primary SO_4^{2-} (46%), NO_3^{-} (86%), V (38%), Ni (70%), and Hg (59%) emissions, despite the large apparent under-estimation of Hg emissions in the inventory. PM_{10} As was emitted mainly by wood (55%) and coal (29%) combustion. Similar to the non-inventoried sources discussed above, specific sources of trace metals are not adequately accounted for.

Comparison with Previous Work

Large variations in PM emission estimates were found in past studies for Delhi (e.g., WHO-UNEP 1992; Bose, 1996; Garg et al., 2001; Gurjar et al., 2004; 2008; Sahu et al., 2011). TSP emissions were estimated to be 58 TPD in 1990 (Sharma et al., 2002), 317 TPD in 1990 (Bose, 1996), and 386 TPD in 1995 (Gurjar et al., 2004). The two high estimates for TSP (i.e., Bose, 1996 and Gurjar et al., 2004, respectively) are more than twice as large as the current estimate of 140 TPD for 2007 PM₁₀. This is reasonable considering that fugitive dust emissions are found in the largest particle size fractions. The vehicular emission factors in NEERI (2010) vary by vehicle age, but they are one to two orders of magnitude lower than those of Gurjar et al. (2004): 0.01-0.07 versus 0.5 g/km for motorcycles and 0.002-0.008 versus 0.5 g per kilometer (km) for gasolinefueled vehicles. Emission factors for diesel-fueled buses are similar: 0.3-2 g/km from NEERI (2010) and 1.15 g/km from Gurjar et al. (2004).

The most recent $PM_{2.5}$ and PM_{10} inventories for Delhi by Sahu *et al.* (2011) are compared with the 2007 PM_{10} emission inventory (NEERI, 2010) in Table 7. PM_{10} emission rates by Sahu *et al.* (2011) are higher (645 TPD) than the 140 TPD estimate in the NEERI (2010) inventory. One contributing factor is the size of the study area: 4550 km² (covering Delhi and surrounding sub-region) in Sahu *et al.* (2011) versus 1500 km² in NEERI (2010), which was



Fig. 1. Percent contributions of diesel-, compressed natural gas (CNG)-, and gasoline-fueled vehicles to PM_{10} mass and chemical species within the vehicular source category (NEERI, 2010).

Table 6. Source contribution of various fuels to daily PM_{10} mass and chemical species in the 2007 city-wide inventory for Delhi, India.

Emission Source		Ре	ercent of	Fotal PM ₁	0 Mass an	d Chemic	cal Specie	s Emissio	ns	
by Fuel Type ^a	PM_{10}	OC	EC	SO_4^{2-}	NO_3^-	Pb	Ni	V	As	Hg
LPG ^b	1.65	1.35	0.59	7.5	0.10	3.8	1.36	1.09	11.2	6.7
CNG^{c}	2.2	2.8	1.56	5.0	9.0	0.58	1.31	1.62	0.12	1.10
Gasoline	2.6	4.5	1.48	5.4	0.68	1.0	0.10	20	0.12	0.00
Diesel	15.1	29	56	22	2.2	1.23	17.5	37	3.8	3.7
Kerosene	0.21	1.98	1.47	1.61	0.05	0.48	0.95	0.94	0.91	1.72
Coal	53	17.1	13.1	46	86	31	70	38	29	59
Wood	25	43	26	12.2	1.45	62	8.3	1.79	55	28.

^a Excluding paved road dust, construction, and non-fuel-specific sources (i.e., waste incineration, open solid waste burning). See Table 3 for different fuel types used in each of the Source Numbers.

^b Liquefied petroleum gas.

^c Compressed natural gas.

Table 7. Comparison of PM emission rates between 2007 NEERI^a (NEERI, 2010) and 2010 Sahu et al. (2011) emission inventories.

			Invent	ory Year		
Base Year	4	2010	2	010	2	2007
Source Category ^b	Sahu et al. (2011) PM _{2.5}		Sahu et al.	(2011) PM ₁₀	NEERI ((2010) PM ₁₀
	Toppes/day	Percent of Total	Toppes/day	Percent of Total	Tonnes/day	Percent of Total
	Tonnes/uay	Emissions	Tonnes/day	Emissions	Tomies/uay	Emissions
Power Plant	7.9	1.4%	30	4.6%	25	18%
Industrial Boilers	45	8.2%	74	11.5%	2.2	1.2%
Residential	51	0.9%	99	15.3%	11.6	8.3%
Paved Road Dust	360	66%	360	56%	77	55%
Vehicle	83	15.2%	82	12.7%	9.7	6.9%
Total Emissions	546		645		140	

^a National Environmental Engineering Research Institute.

^b For NEERI (2010) see Table 3 for details on Power Plants (Source Numbers 1 and 2), Industrial Boilers (Source Numbers 3 and 4), Paved Road Dust (Source Number 20), and Vehicle (Source Numbers 21–30) emission rates. Residential includes domestic cooking and heating sources (Source Numbers 5–8).

focused more on the city center. Nonetheless, Sahu *et al.* (2011) estimate PM_{10} emissions over 8 times higher for vehicular and residential sources and over 30 times higher for industrial sources.

Speciated Emission Inventory Uncertainties

It is clear that PM emission inventories vary widely from study to study. Annual emission estimates depend on the representativeness of specified emission factors and activity levels. There are no standards for estimating activity levels, and consistency must be established among independent studies and comparisons need to be made between measured ambient concentrations and those estimated from models in the inventories. Speciated PM emissions also depend on the representativeness of the chemical source profiles. Even in the most advanced air quality and source apportionment studies, source profiles are often non-representative or inaccurate (Watson et al., 2008). This is evident for vehicular sources, emissions from which can represent a mixture from thousands if not millions of different vehicles. In the 2007 study, local source profiles were developed based on a limited number of tests (e.g., four post-2000 vintage dieselfueled heavy-duty commercial vehicles [ARAI, 2009] and two coal-fired power plants [Sethi and Patil, 2008a]). Source profiles should be continuously improved upon with more vehicle tests under different driving conditions covering a broad range of fuels and engine types and ages (e.g., Wang et al., 2012).

Although several area and vehicular source emission factors were specifically measured in India, with some in Delhi, the 2007 inventories (NEERI, 2010) rely heavily on U.S. AP-42 emission factors (U.S.EPA, 2006). Most of these emission factors result from laboratory testing of only a few individual emitters, whereas more real-world emission factors require measurements for the actual hardware, fuels, and operating conditions specific to the study region (Watson et al., 2012). Domestic cooking activities in Delhi constitute another major uncertainty, as LPG is the dominant fuel near the city center, while greater amounts of wood and kerosene are used in the lower-income areas at the periphery of the city. Fugitive dust emissions are based on silt loadings, and this activity indicator is not considered accurate in North America for PM₁₀ or PM₂₅ size fractions (Watson et al., 2000). As noted, uncertainties of emission inventories and source characterization should be evaluated. A weight-of-evidence approach (U.S. EPA, 2007) should be used to reconcile differences between source modeling and ambient measurements in future studies.

CONCLUSIONS

Paved road dust, construction activities, power plants, domestic cooking and vehicles are the main sources of PM_{10} in Delhi. Ground-level emissions of toxic species are the most significant sources in terms of exposure and impacts on human health. For the selected toxic metals, paved road dust and domestic cooking are the principal ground-level sources of PM_{10} Pb, Ni, and Hg while vehicle exhaust is the main source of V. Residential wood combustion for cooking and heating is the dominant source of As.

Paved road dust dominates PM_{10} emissions (55%) and is the main source of OC (33%) and EC (34%). Dust suppression should be an important component of air quality remediation efforts in Delhi. Vehicles are the next most important source of EC (33%). While conversion to cleaner fuels like CNG and movement towards a newer fleet will lower vehicle emissions, an efficient public transport system that minimizes the use of personal vehicles, especially two-wheelers, is the key to improving air quality in Delhi. This would also reduce traffic volume and, in turn, decrease re-suspension of road dust.

Efforts should be made to use natural gas for domestic cooking and to eliminate other cooking fuels (e.g., coal, kerosene, and wood). Estimated speciated PM_{10} emissions need to be further refined by determining more representative source profiles for PM_{10} from specific sources that emit toxic chemical species. It is also necessary to continue ambient measurements of $PM_{2.5}$, PM_{10} , and their major chemical components in order to establish a long-term database for evaluating the effectiveness of pollution control measures.

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