



Characterization of PM_{2.5} Source Profiles for Traffic and Dust Sources in Raipur, India

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ABSTRACT

This paper describes results from a detailed source profile characterization study conducted in Raipur, India to prepare source profiles for traffic and dust-related sources. A companion paper has been published with results for a range of other combustion sources. PM_{2.5} samples were analyzed for mass, elements (Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, Pb, S, Sb, Se, V, Zn), ions (Na⁺, K⁺, Mg²⁺, Ca²⁺, NH₄⁺, Cl⁻, F⁻, NO₃⁻, SO₄²⁻) and carbonaceous fractions (OC and EC). All dust profiles were dominated by crustal elements (Al, Ca, Fe and Mg), while carbonaceous species (OC and EC) were most abundant in vehicular emission profiles. Trace element fraction was found to be significantly higher in vehicular exhaust compared to the resuspended dust. Remarkably, sulphur abundance was observed to be several-fold higher in vehicular emission profiles than resuspended dust profiles. Al and Ca were identified as reliable markers for resuspended dust while V, Pb and EC were identified as markers for vehicular exhaust.

Keywords: PM_{2.5}; Source profiles; Source markers; Traffic; Dust.

INTRODUCTION

Air quality is one of the key environmental and public health concerns in urban areas, and quantitative estimates of source contributions are important for policy design (Labban *et al.*, 2004). Receptor models such as the chemical mass balance (CMB) model use ambient concentrations of particulate matter (PM) and source profiles to quantify source contributions. Source profiles, defined as “fractional mass abundances and uncertainty values of chemical species relative to primary PM emission sources” (Watson *et al.*, 2001), are typically generated for different source types, and can be used as input for the CMB model as well as for comparison with multivariate model factors, and for calculation of emission inventories (Watson *et al.*, 2001; Zannetti, 2005). The SPECIATE database maintained by

the USEPA has a large collection of US-specific profiles, and several studies have reported source profiles elsewhere including India (Gupta *et al.*, 2007; Patil *et al.*, 2013), China (Ho *et al.*, 2003; Zhao *et al.*, 2006; Cao *et al.*, 2008; Kong *et al.*, 2010; Kong *et al.*, 2011) and Europe (Amato *et al.*, 2009; Pant *et al.*, 2014). The profiles differ across source types and are influenced by a range of factors. In case of vehicle emission profiles, factors include operating conditions, fuel type, combustion and emission control technologies and in case of soil material; local geology and geographic location introduces variation (Watson and Chow, 2001; Yatkin and Bayram, 2008). Hence, it is important to develop locally-representative source profiles in order to quantify source contributions correctly. However, source apportionment studies based on the CMB model from India typically use the SPECIATE profiles due to limited availability of locally derived source profiles for PM_{2.5} fraction and this can introduce large uncertainties in the source contribution estimates (Karar and Gupta, 2007; Chakraborty and Gupta, 2010; Matawle *et al.*, 2014; Patil *et al.*, 2013). In a detailed review on receptor modeling studies in India, Pant and Harrison (2012) highlighted the need for development of local source profiles. Although vehicular exhaust and resuspended

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dust are two of the major sources of particulate matter in India, there are a limited number of studies on chemical characterization studies of the sources (Chowdhury *et al.*, 2007; Srivastava and Jain, 2007; Balakrishna and Pervez, 2009; Khare and Baruah, 2010). As part of a national study on source apportionment, a set of 27 emission sources of PM₁₀ and PM_{2.5} were characterized across six major cities of India, and results from the study highlighted significant variations in species' concentrations across the locations and source types (Patil *et al.*, 2013). However, while this study produced a range of source profiles for PM₁₀, the number of profiles for PM_{2.5} was relatively limited. In addition to source characterization, previous studies have used other approaches including use of diagnostic tools such as elemental ratios and enrichment factors to identify chemical source markers for PM sources (Artidsoglou and Samara, 2005; Han *et al.*, 2006; Han *et al.*, 2007; Tsai *et al.*, 2007; Cao *et al.*, 2008; Cheng *et al.*, 2010; Kong *et al.*, 2011; Cesari *et al.*, 2012). The key objective of this study was to prepare PM_{2.5} source profiles for resuspended dust and vehicular exhaust emission sources for Raipur (India), and a companion paper on other profile categories has recently been published (Matawle *et al.*, 2014).

METHODOLOGY

Study Area and Sampling

The study area, Raipur is the capital city of Chhattisgarh, India (21°14'22.7"N, 81°38.1"E); with a total population of 1,010,087 (Census, 2011). A detailed description of the study site is provided in Matawle *et al.* (2014). In this study, PM_{2.5} emissions from two major source categories, i.e., vehicular exhaust and dust were characterized. Each source category was divided into four subcategories (Table 1).

Paved and unpaved road dust samples were collected by sweeping 0.5 to 1 kg of the material using a clean plastic pan and brush from the surface and stored in zipped polyethylene bags (Table 1). The soil samples were collected using procedures in earlier reported studies (Watson and Chow, 2001; Watson *et al.*, 2001; Kong *et al.*, 2011; Patil *et al.*, 2013). Using the method described by Chow *et al.* (1994), samples were dried, sieved (Tyler 400-mesh screen) and resuspended in a laboratory chamber and sampled through PM_{2.5} inlets at an average flow rate of 5 L min⁻¹ (MINIVOL Ver. 4.2, AirMetrics) for 5 to 10 minutes to ensure sufficient deposition onto quartz fiber filters (referred to as QFF hereafter) (1851-047, Whatman UK). Based on traffic count surveys conducted during the study and other published data (CPCB, 2008b; Road Transport Year Book, 2011), four major vehicle categories were selected- (i) two wheelers, (ii) three wheelers, (iii) passenger vehicles and (iv) heavy duty vehicles (HDVs). Samples were collected on QFFs using a PM_{2.5} sampler inlet placed in the hot exhaust point of the vehicles using previously used methods (Watson *et al.*, 1994, Watson and Chow 2002; Chow *et al.*, 2004; Patil *et al.*, 2013). In order to represent typical emission characteristics across different vehicle age classes, vehicles manufactured between 2000 and 2010 were selected. The medium accelerator operating condition was adjusted during

Table 1. Description of PM_{2.5} sources emission profiles Raipur, India.

S.N. Profile ID	Source Name	Description	Sampling methodology
resuspended dust emission sources			
1.	Soil	Natural soil dust	Chamber re-suspension sampling
2.	CD	Civil construction dust	Chamber re-suspension sampling
3.	PRD	Paved road dust	Chamber re-suspension sampling
4.	UPRD	Unpaved road dust	Chamber re-suspension sampling
Vehicular emission sources			
1.	2WVG	Two wheelers vehicles (gasoline)	In-plume sampling
2.	3WVD	Three wheeler vehicles (diesel)	In-plume sampling
3.	4WVD	Four wheeler vehicles (diesel)	In-plume sampling
4.	DHDV	Heavy duty vehicles (diesel)	In-plume sampling
		Composite of 5 samples collected from non-agricultural area outside of Raipur City (non-agricultural area was selected so as to not have any influence from fertilizer/pesticide use)	Chamber re-suspension sampling
		Composite of 5 samples collected from a construction site located in Raipur city	Chamber re-suspension sampling
		Composite of 5 samples collected from surface of paved road of Raipur city	Chamber re-suspension sampling
		Composite of 5 samples collected from surface of unpaved road outside of Raipur City	Chamber re-suspension sampling
		Composite of 5 samples collected from petrol driven 2-wheelers	In-plume sampling
		Composite of 5 samples collected from diesel driven 3-wheeler passenger auto rickshaws	In-plume sampling
		Composite of 5 samples collected from diesel driven 4-wheeler personal cars	In-plume sampling
		Composite of 5 samples collected from diesel driven heavy duty trucks	In-plume sampling

sampling for 30–60 minutes time cycle.

The sampled filters were weighed before and after sampling under controlled environment (RH- 35% \pm 1% and temperature- 22.0°C \pm 1°C) using a six digit electronic balance (Denver Instrument, Model TB-2150, Germany) with \pm 10 μ g precision. After weighing, the filters were stored at -5°C until chemical analysis.

Chemical Analysis

The PM_{2.5} samples were analyzed for 32 chemical species including 21 elements- Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, Pb, S, Sb, Se, V, Zn; 9 water-soluble ions- Na⁺, K⁺, Mg²⁺, Ca²⁺, NH₄⁺, Cl⁻, F⁻, NO₃⁻, SO₄²⁻ and organic carbon (OC) and elemental carbon (EC). Each QFF was cut into four equivalent portions. One portion was used for acid digestion (1:3 H₂O₂/HNO₃) and the extracts were used to analyse elemental species using a graphite furnace/cold vapour equipped atomic absorption spectrophotometer (AAS) (iCE3500 Model, Thermo Fisher). Another portion was used for water extraction (for ion analysis) by ultrasonication in a 20 mL vial using 10 mL distilled water (18 M Ω) for one hour as well followed by mechanical shaking for one hour. The extracts were used to analyse cations and anions using ion chromatography (Model 2000, Dionex). SO₄²⁻ measurements in both water extracts and digested samples of PM_{2.5} was carried out to quantify sulphur content of PM_{2.5} by subtracting the SO₄²⁻ (digested) from SO₄²⁻ (water extracts) (Gurugubelli *et al.*, 2013). NH₄⁺ was determined spectrophotometrically (1305E Model, Systronic) by following standard indophenol blue method at 620 nm wavelengths (Harrison and Perry, 1986). The samples were analysed for OC and EC using a Sunset semi-continuous thermal/optical carbon analyser (Sunset Laboratory, Model 4L, USA) with the NIOSH-5040 protocol (NIOSH, 1999). All samples were corrected for field blanks, and chemical abundances were calculated as “fractional mass abundance of measured chemical species relative to primary PM_{2.5} mass” as described in Chow *et al.* (2003). Uncertainty of source profile abundances was estimated as the standard deviation of the average from five source tests. Details of the methods used, and the limits of detection are presented in Matawle *et al.* (2014).

RESULTS AND DISCUSSION

Source Profiles for Resuspended Dust

Four different dust sources were characterized in this study including soil, construction dust (CD), paved road dust (PRD) and unpaved road dust (UPRD). Table 2 describes the various PM_{2.5} source profiles. These profiles were found to be dominated by Al, Ca, Fe, Mg, OC and EC although major inorganic constituents (Ca, Fe, and Al) showed a higher degree of variation (43–99%) across the selected profiles. Total carbon (TC, described as sum of OC and EC) accounted for 14–40.3% of the PM mass while crustal fraction was found to account for 43.5–83.8% of the PM mass across profiles. Results observed in the current study were comparable to those reported earlier (Chow *et al.*, 1994; Watson *et al.*, 2001; Amato *et al.*, 2009; Patil *et al.*,

2013). The soil profile showed higher abundances of Al and Ca compared to other major inorganic constituents. K was found to be two-fold higher than soluble K⁺ ion with a K⁺/K ratio 0.40; comparable to those reported earlier (0.1–0.5) (Watson *et al.*, 2001; CPCB, 2008a; Kong *et al.*, 2014) and this is contrary to contribution from the biomass emissions where the typical K⁺/K ratio is between 0.87–0.90 (Chow *et al.*, 2004; Watson *et al.*, 2001). The abundance of OC in soil was found to be similar to previously reported values for Indian conditions (CPCB, 2008a) but higher than those reported in China (Kong *et al.*, 2014). The OC/TC ratio for the soil profile was observed to be 0.69 for the soil profile and the value was broadly comparable to those reported earlier (0.64–0.99) (Ho *et al.*, 2003; Chow *et al.*, 2004; Gupta *et al.*, 2007). In comparison, the highest OC/TC ratio (0.96) was observed for the CD profile. In case of the CD profile, Ca was observed to be the most abundant species with the lowest uncertainty value (21.4 \pm 1.78%). Ca has previously been reported as a source marker for construction activity (Yatkin and Bayram, 2008; Pant and Harrison, 2012). Al, Fe, OC and EC were found comparatively lower than those found in the soil profile, whereas Na and Mg were observed to be two times higher than the soil profile. The EC abundance was observed to be significantly lower with higher variability.

The PRD and UPRD profiles were dominated by Ca, Fe, Mg, Ca²⁺, OC, and EC. Interestingly, Ca²⁺/Ca ratio was found to be 0.24 and 0.22 for PRD and UPRD, respectively; indicating a crustal origin, because soluble content of elements are reported to be abundant in emissions resulting from combustion activities (Volkovic, 1983; Watson *et al.*, 2001). Fe was found to be more abundant in the UPRD profile with an average value of 12.1 \pm 0.86% compared to the PRD profile (9.78 \pm 0.70%). This could be attributed to the local geological chemistry and presence of a large number of iron processing industries in the study area. Both OC and EC were found to be higher in the PRD compared to UPRD profile, which is expected since exhaust particles can deposit on PRD. Additionally, carbonaceous matter (OC+EC) was approximately 1.5 times higher in road dust profiles (PRD and UPRD) compared to those found in similar source profiles reported earlier for different Indian regions (Patil *et al.*, 2013). Pb, Zn and SO₄²⁻ abundances were found higher in PRD profile than UPRD profiles and similar to those found in earlier profiles in other Indian cities (CPCB, 2008a). The higher occurrence of Pb, Zn and SO₄²⁻ in both PRD and UPRD indicates contribution from traffic emissions and re-suspension of asphalt particles (Watson *et al.*, 2001; Samara, 2005). Sulphur content of the petrol and diesel used in central India is 150 and 350 ppm respectively, which could potentially contribute to higher sulphate concentration in the road dust profiles. Lead content in petrol in India is reported to be approximately 15 ppm (SIAM, 2015).

The PM_{2.5} mass was reconstructed based on the measured chemical species using the mass balance equation IMPROVE18 described by Chow *et al.* (2012). IMPROVE18 equation was used to perform quality control parameter on PM_{2.5} datasets by comparison between gravimetrically measured mass and sum of the all speciated chemical species concentrations.

Table 2. PM_{2.5} sources profiles for vehicular sources (expressed as weight percent by mass).

Species	2WVG	3WVD	4WVD	DHDV
Al	0.00116 ± 0.00467	0.00401 ± 0.00553	0.00590 ± 0.00377	0.00425 ± 0.00676
As	0.00033 ± 0.00160	0.00017 ± 0.00021	0.00018 ± 0.00030	0.00052 ± 0.00020
Ca	0.08364 ± 0.09556	0.13064 ± 0.08961	0.01070 ± 0.05465	0.03372 ± 0.05677
Cd	0.00012 ± 0.00036	0.00008 ± 0.00007	0.00024 ± 0.00145	0.00002 ± 0.00010
Co	0.00027 ± 0.00010	0.00009 ± 0.00008	0.00047 ± 0.00010	0.00031 ± 0.00010
Cr	0.00374 ± 0.00160	0.00176 ± 0.00414	0.00043 ± 0.00212	0.00138 ± 0.00200
Cu	0.07868 ± 0.03810	0.02376 ± 0.00591	0.02697 ± 0.00513	0.02230 ± 0.00230
Fe	0.17517 ± 0.09578	0.34780 ± 0.01556	0.27920 ± 0.00284	0.12522 ± 0.01240
Hg	0.00406 ± 0.01310	0.00068 ± 0.00071	0.00163 ± 0.00030	0.00275 ± 0.00600
K	0.00289 ± 0.00126	0.01667 ± 0.00534	0.00223 ± 0.00187	0.00264 ± 0.00090
Mg	0.00509 ± 0.00361	0.02735 ± 0.00225	0.00283 ± 0.00197	0.00306 ± 0.00249
Mn	0.00592 ± 0.00237	0.00280 ± 0.00212	0.00291 ± 0.00621	0.00271 ± 0.01160
Mo	0.00409 ± 0.01090	0.00291 ± 0.00735	0.00131 ± 0.00511	0.00222 ± 0.00410
Na	0.67308 ± 2.57500	0.28844 ± 0.32535	0.79874 ± 0.04660	3.41604 ± 0.63310
Ni	0.00895 ± 0.02240	0.00178 ± 0.00076	0.00572 ± 0.00312	0.00599 ± 0.01300
Pb	0.06127 ± 0.00290	0.38000 ± 0.01200	0.50000 ± 0.00112	0.80000 ± 0.02000
S	1.97260 ± 0.05379	1.64250 ± 0.03142	1.66600 ± 0.04700	2.36379 ± 0.02966
Sb	0.03400 ± 0.00302	0.02240 ± 0.01220	0.01400 ± 0.00660	0.02210 ± 0.01002
Se	0.12300 ± 0.01240	0.40000 ± 0.02000	0.06000 ± 0.01120	0.80000 ± 0.02000
V	0.00836 ± 0.00624	0.00138 ± 0.00171	0.00189 ± 0.00094	0.00491 ± 0.00680
Zn	0.51284 ± 0.41251	0.81470 ± 0.11730	0.61171 ± 0.00557	0.92522 ± 0.01496
F ⁻	0.17825 ± 0.03285	0.00023 ± 0.00012	0.08883 ± 0.00728	0.01245 ± 0.00120
Cl ⁻	0.17491 ± 0.06846	0.73487 ± 0.26485	0.59327 ± 0.02813	1.66916 ± 0.13607
NO ₃ ⁻	0.00246 ± 0.00250	0.00620 ± 0.00254	0.11762 ± 0.08125	0.00213 ± 0.00025
SO ₄ ²⁻	0.13562 ± 0.06983	0.88586 ± 0.08541	0.98512 ± 0.01727	1.70472 ± 0.13023
Na ⁺	0.66830 ± 1.10200	0.11393 ± 0.05100	0.29651 ± 0.01412	0.43976 ± 0.06260
NH ₄ ⁺	0.09146 ± 0.00743	0.78760 ± 0.03410	0.13610 ± 0.04600	1.68230 ± 0.45300
K ⁺	0.00218 ± 0.00089	0.00585 ± 0.00093	0.01037 ± 0.00037	0.00240 ± 0.00090
Ca ²⁺	0.07919 ± 0.03521	0.11942 ± 0.04254	0.00848 ± 0.00104	0.03142 ± 0.01100
Mg ²⁺	0.00470 ± 0.00151	0.02200 ± 0.00400	0.00250 ± 0.00120	0.00340 ± 0.00010
OC	55.01420 ± 6.88900	47.85230 ± 9.26150	35.16210 ± 5.66478	29.05820 ± 1.83906
EC	0.93430 ± 0.57390	5.61050 ± 0.63790	13.15830 ± 6.08020	21.35270 ± 0.17372
Sum%	60.29047 ± 7.58692	59.98747 ± 6.59812	54.23438 ± 5.81643	64.02080 ± 9.42822

$$[\text{PM}_{2.5}] = 1.375 \text{SO}_4^{2-} + 1.29 \text{NO}_3^- + 1.8 \text{OC} + \text{EC} + (2.2 \text{Al} + 2.49 \text{Si} + 1.63 \text{Ca} + 1.94 \text{Ti} + 2.42 \text{Fe}) + 1.8 \text{Cl}^- \quad (1)$$

According to Chow *et al.* (2015), several factors influences mass reconstruction result are (i) sampling artifacts, (ii) unmeasured chemical species, (iii) OC multiplier factor, (iv) carbon analysis method, (v) NH₄⁺ and NO₃⁻ volatilization, and (vi) water uptake on filter paper at different equilibrated RHs. The mass reconstruction results were explained 58–75% of the observed mass for dust profiles. It is important to highlight that Si and Ti were not measured during this study, and this could have resulted in lower estimated values for the crustal fraction. Previous studies indicate that elements such as Si (SiO₂) and Ti (TiO₂) can contribute up to 30% of soil or dust mass (Watson *et al.*, 2003; Chow *et al.*, 2004).

Vehicular Exhaust Profiles

The vehicular source profiles (Table 3) were found to be dominated (> 79–92% of measured mass) by carbonaceous matter. OC was found to be the most abundant species in all vehicular profiles with values ranging from 48.3 ± 11.7% in 4WVD to 55.9 ± 7.46% in 2WVG profiles. In comparison,

EC was found in the range of 0.93 ± 0.57% in 2WVG to 21.4 ± 0.17% in HDVD profile. The significantly lower EC values observed in 2WVG profile is due to the lower EC content in petrol-driven vehicles compared to diesel-driven vehicles and similar results were reported in other Indian profiles (CPCB, 2008b). The OC/TC ratios were found in the range of 0.57 (HDVD) to 0.98 (2WVG) profiles with an average value of 0.79 ± 0.18 across vehicular profiles. The values are comparable to those reported earlier in USA (0.39–0.80) (Watson *et al.*, 2001) and India (0.64–0.94) (CPCB, 2008b). SO₄²⁻/S ratio for petrol vehicles and all three types of diesel vehicles (3WVD, 4WVD and DHDV) showed SO₄²⁻/S ratio between 0.55 and 0.73, whereas the ratio value was 0.06 for petrol 2WVG. The higher sulphur abundance (in range 1.64–2.36% for Raipur) can be attributed to the use of high sulphur fuel in parts of India. Pb and Zn abundances were found to be 0.06 ± 0.00% to 0.80 ± 0.02% and 0.51 ± 0.41% to 0.93 ± 0.01% across vehicular exhaust profiles. Overall, 79–101% of the total PM_{2.5} mass was reconstructed using this profile based on calculations detailed in Eq. (1) (Chow *et al.*, 2012). The mass reconstruction results for vehicular exhaust profiles were comparatively higher

Table 3. PM_{2.5} sources profiles for resuspended dust emission sources (expressed as weight percent by mass).

Species	Soil	CD	PRD	UPRD
Al	1.92047 ± 1.05010	0.91270 ± 0.26421	0.20399 ± 0.01492	0.21865 ± 0.01573
As	0.00094 ± 0.00770	0.00010 ± 0.00010	0.01035 ± 0.00832	0.00584 ± 0.00381
Ca	9.15143 ± 0.80585	21.38090 ± 1.77580	11.23563 ± 0.79679	10.86861 ± 0.76977
Cd	0.00015 ± 0.00031	0.00052 ± 0.00063	0.00640 ± 0.00091	0.00138 ± 0.00031
Co	0.01121 ± 0.00800	0.00227 ± 0.00635	0.00316 ± 0.00227	0.00142 ± 0.00102
Cr	0.37484 ± 0.01894	0.00235 ± 0.00255	0.10474 ± 0.00806	0.00703 ± 0.00078
Cu	0.22389 ± 0.09086	0.02567 ± 0.00543	0.02292 ± 0.00234	0.01447 ± 0.00134
Fe	6.96727 ± 0.42042	4.82199 ± 0.58193	9.78430 ± 0.69413	12.08730 ± 0.85593
Hg	0.01272 ± 0.04928	0.00435 ± 0.01637	0.01001 ± 0.00269	0.01193 ± 0.00167
K	1.48782 ± 0.08008	0.15495 ± 0.08110	0.67737 ± 0.16448	0.46884 ± 0.09498
Mg	1.18924 ± 0.06052	2.04071 ± 0.03368	1.09803 ± 0.07866	1.49239 ± 0.10599
Mn	0.47246 ± 0.05713	0.20625 ± 0.02515	0.10664 ± 0.00776	0.09697 ± 0.00696
Mo	0.00109 ± 0.01032	0.00548 ± 0.01587	0.00560 ± 0.00261	0.00272 ± 0.00119
Na	0.93572 ± 0.04575	2.72404 ± 0.32500	0.42765 ± 0.03829	0.24249 ± 0.02060
Ni	0.10264 ± 0.03496	0.02584 ± 0.00310	0.05615 ± 0.01766	0.01745 ± 0.00744
Pb	0.00165 ± 0.00123	0.00053 ± 0.00045	0.15370 ± 0.02398	0.00810 ± 0.00678
S	0.07327 ± 0.01070	0.00475 ± 0.00154	0.09589 ± 0.00815	0.01452 ± 0.00425
Sb	0.00370 ± 0.00193	0.00073 ± 0.00180	0.01137 ± 0.00817	0.01079 ± 0.00400
Se	0.02161 ± 0.00137	0.00037 ± 0.00527	0.02735 ± 0.00849	0.01374 ± 0.00391
V	0.01114 ± 0.00764	0.01885 ± 0.00128	0.03467 ± 0.00604	0.01486 ± 0.00268
Zn	0.61525 ± 0.07608	0.25373 ± 0.02253	0.07835 ± 0.00585	0.00786 ± 0.00071
F ⁻	0.00770 ± 0.00470	0.00099 ± 0.00025	0.53706 ± 0.04132	0.04959 ± 0.00519
Cl ⁻	1.36598 ± 0.56518	0.02779 ± 0.01700	0.63982 ± 0.22715	0.15709 ± 0.11285
NO ₃ ⁻	0.71134 ± 0.02834	0.01583 ± 0.06534	0.06355 ± 0.04565	0.03467 ± 0.02490
SO ₄ ²⁻	1.04198 ± 1.57542	0.00568 ± 0.00402	2.96455 ± 0.22934	0.46653 ± 0.04452
Na ⁺	0.20680 ± 0.00641	0.00002 ± 0.00001	0.37913 ± 0.27237	0.07612 ± 0.11882
NH ₄ ⁺	0.10620 ± 0.10179	0.01518 ± 0.01019	0.16064 ± 0.11540	0.07302 ± 0.05246
K ⁺	0.60606 ± 0.25142	0.01188 ± 0.01052	0.28861 ± 0.20734	0.13120 ± 0.09425
Ca ²⁺	1.73216 ± 0.33326	0.33330 ± 0.08241	2.69896 ± 0.51814	2.39488 ± 0.30959
Mg ²⁺	0.02695 ± 0.03326	0.08308 ± 0.03112	0.14940 ± 0.10599	0.96483 ± 0.49764
OC	12.54823 ± 0.97433	5.12300 ± 2.89701	12.62781 ± 0.63139	6.55793 ± 0.32790
EC	5.52860 ± 0.18311	0.18345 ± 0.42730	3.82685 ± 0.19134	2.57670 ± 0.12884
Sum%	44.88854 ± 4.01251	37.95901 ± 3.86152	44.97457 ± 3.62154	35.52292 ± 3.28561

than dust profiles due to lower abundances of unmeasured elemental species associated to vehicular exhaust PM_{2.5}.

Abundances across Source Profiles

Each composite source profile developed in this study consists of average fractional mass abundances and uncertainty (represented as standard deviation) for 32 chemical species. For the resuspended dust source profiles (soil, CD, PRD and UPRD), the measured mass was 44.9 ± 4.01%, 37.96 ± 3.86%, 44.97 ± 3.62% and 35.5 ± 3.29% respectively. On the other hand, for the vehicular source profiles (2WVG, 3WVD, 4WVD and DHDV), the values were 60.3 ± 7.59%, 59.99 ± 6.60%, 54.2 ± 5.82% and 64.0 ± 9.43% respectively. A greater proportion of the measured mass was accounted for vehicular profiles compared to the resuspended dust profiles and this can be due to the higher abundances of the unmeasured oxygen in the form of oxides for elements such as Si, Ti and P and water content (Ho *et al.*, 2003; Samara *et al.*, 2003; Samara, 2005).

In order to compare the profiles, chemical species were further grouped into five classes: (i) crustal elements (Al, Ca, Fe, K, Mg, Mn and Na), (ii) ions (F⁻, Cl⁻, NO₃⁻, SO₄²⁻,

Na⁺, K⁺, Ca²⁺ and Mg²⁺), (iii) trace elements (As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, S, Sb, Se, V, and Zn), (iv) OC and (v) EC as described in earlier reports (Kong *et al.*, 2011) and compared graphically in Fig. 1. The crustal element fraction was the most abundant in resuspended dust profiles, with average mass of 19.6 ± 1.90%, 31.8 ± 2.96%, 20.0 ± 0.69% and 21.9 ± 0.85%, for soil, CD, PRD and UPRD respectively while the OC and EC fractions were found to be more abundant in vehicular profiles with concentrations ranging from 29.1% to 55.0%. The trace element fraction was found in significantly higher concentrations in vehicular profiles compared to the resuspended dust profiles with abundances of 2.81 ± 0.58%, 3.29 ± 0.21%, 2.89 ± 0.09% and 4.95 ± 0.13% for 2WVG, 3WVD, 4WVD and HDVD respectively in comparison with 1.45 ± 0.32%, 0.35 ± 0.08%, 0.62 ± 0.11% and 0.13 ± 0.04% for soil, CD, PRD and UPRD respectively. The ion fraction had comparable abundance across both sets of profiles ranging from 0.49 ± 0.22% for CD to 7.88 ± 1.76% for PRD.

Comparisons among PM_{2.5} Source Profiles

Similarities and differences among the developed PM_{2.5}

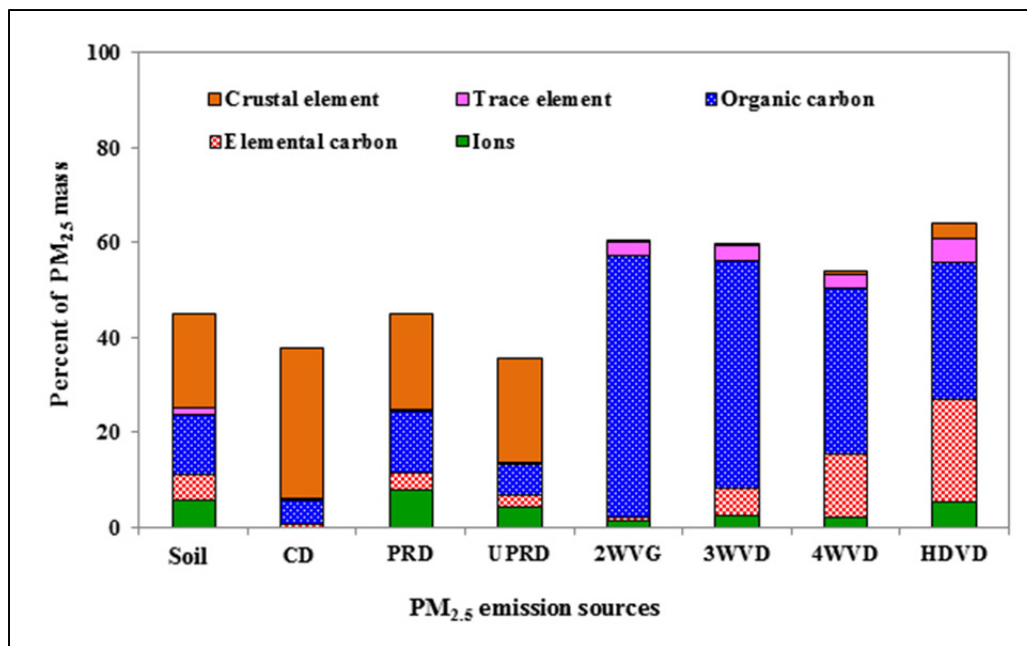


Fig. 1. Chemical composition of the exhaust and dust source profiles.

source profiles were estimated by coefficient of divergence (COD) statistic. The coefficient of divergence is a self-normalizing statistical parameter and can be used to measure the spread of the data points. It is typically used to estimate spatial variability of pollutant concentrations and is calculated as follows (Wongphatarakul *et al.*, 1998; Zhang and Friedlander, 2000):

$$COD_{jk} = \sqrt{\frac{1}{p} \sum_{i=1}^j \left(\frac{X_{ij} - X_{ik}}{X_{ij} + X_{ik}} \right)^2} \quad (2)$$

where j and k refer to profiles 1 and 2, p refers to the number of investigated components, and X_{ij} and X_{ik} represented the average mass concentrations of chemical component i for j and k .

Values approaching zero indicate similarity while values approaching one indicate dissimilarity. According to Wongphatarakul *et al.* (1998), the COD values ≤ 0.269 shown similarity between two different samples. Recent studies have also used this parameter to compare source profiles (Kong *et al.*, 2011). In this study, the COD values were calculated for 19 different PM_{2.5} source profiles dataset combinations [8 profiles in this study and 11 profiles reported by Matawle *et al.* (2014)] for the Raipur-Bhilai region (India). Results are presented in Table 4. The COD values ranged from 0.48 to 0.84 for all profile combinations. COD values for all PM_{2.5} source profile combinations indicated that these profiles were significantly different from each other. Further, the measured species in developed profiles were grouped in four abundance ranges to compare those grouped in earlier reported profiles (Table S1) (Watson *et al.*, 2001; Chow *et al.*, 2004; CPCB, 2008a, b). All source profiles have shown good similarities with earlier reported profiles. In case of

major abundant species ($> 10\%$), carbonaceous fractions (OC and EC) is found to be similar in current and reported profiles of vehicular exhaust and PRD. The CD profile was dominated by calcium, whereas the UPRD files had high concentrations of Ca and Fe. Significant differences have been observed in the species grouped for 1–10% abundances compared to those grouped in reported profiles of Indian origin and others. Higher number of species in 1–10% abundances in the developed profiles compared to reported profiles might be due to urban-industrial environmental character of the study area. Trace elements were distributed from $< 0.1\%$ to 1% abundance ranges; similar to those found in reported profiles. But significant dissimilarities have been observed in 0.1–1% and 1–10% abundant chemical species (most of ions and major crustal elements) with different distribution pattern for each profile.

Source Signature and Fingerprints

Source signatures of particles are mostly described by specific size distribution, specific suite of elements and specific ratios of compounds, elements or isotopes (Mitra *et al.*, 2002). Elemental ratios have previously been used as diagnostic tools for a qualitative estimation of possible sources (Arditsoglou and Samara, 2005; Kong *et al.*, 2011). For example, Cu/Sb and Cu/Zn ratios have been used for traffic emissions whereas V/Ni ratio has been used to assess emissions from both shipping emissions and oil burning (Pey *et al.*, 2010). Arditsoglou and Samara (2005) used the Zn/Pb ratio in the ranges of 0.3–0.4 as a fingerprint of automotive emissions, whereas 1.2 was assigned to oil combustion emissions. In another study, Mn/V was used to distinguishing between oil burning ($\ll 1$) and coal burning ($\gg 1$) emission sources (Mitra *et al.*, 2002). Amato *et al.* (2009) proposed a characteristic brake wear ratio of 7.0 ± 1.9 in Europe. In this study, elemental ratios of Mn/V, Cu/Sb,

Table 4. Coefficient of divergence matrix for similarity comparison among various PM_{2.5} profiles for Raipur-Bhilai region, Chhattisgarh.

	2WVG	3WVD	4WVD	DHDV	Soil	CD	PRD	UPRD	MSWB	RSFS	RKS	LPGS	CPI	FEMNI	SPI	CTPP	SRM	BKP	EAW
2WVG	0.00																		
3WVD	0.54	0.00																	
4WVD	0.51	0.52	0.00																
DHDV	0.50	0.50	0.46	0.00															
Soil	0.77	0.78	0.74	0.78	0.00														
CD	0.77	0.79	0.81	0.82	0.70	0.00													
PRD	0.76	0.78	0.74	0.80	0.55	0.71	0.00												
UPRD	0.75	0.80	0.75	0.82	0.61	0.66	0.48	0.00											
MSWB	0.70	0.72	0.71	0.74	0.63	0.75	0.65	0.67	0.00										
RSFS	0.74	0.72	0.72	0.74	0.65	0.80	0.66	0.76	0.54	0.00									
RKS	0.77	0.75	0.71	0.70	0.79	0.90	0.77	0.82	0.81	0.79	0.00								
LPGS	0.72	0.75	0.67	0.68	0.70	0.79	0.65	0.73	0.68	0.76	0.66	0.00							
CPI	0.71	0.78	0.73	0.76	0.69	0.62	0.70	0.70	0.77	0.77	0.84	0.77	0.00						
FEMNI	0.78	0.82	0.78	0.80	0.52	0.68	0.60	0.59	0.65	0.69	0.81	0.72	0.65	0.00					
SPI	0.78	0.83	0.77	0.83	0.56	0.66	0.57	0.58	0.64	0.63	0.81	0.76	0.70	0.48	0.00				
CTPP	0.74	0.79	0.75	0.80	0.58	0.76	0.57	0.67	0.64	0.63	0.79	0.70	0.70	0.58	0.57	0.00			
SRM	0.79	0.77	0.68	0.72	0.60	0.72	0.57	0.59	0.66	0.67	0.79	0.72	0.68	0.62	0.63	0.67	0.00		
BKP	0.73	0.78	0.74	0.78	0.64	0.78	0.54	0.64	0.60	0.61	0.70	0.62	0.75	0.62	0.65	0.58	0.66	0.00	
EAW	0.76	0.81	0.79	0.82	0.68	0.73	0.62	0.67	0.67	0.71	0.81	0.79	0.73	0.63	0.61	0.62	0.75	0.67	0.00

Bold and italic letters indicated COD values for PM_{2.5} source profiles reported by Matawle et al. (2014).

As/V, V/Ni, Zn/Pb, Zn/Cd, Cu/Zn, Cu/Cd, and Cu/Pb were calculated and compared with previous reported studies to investigate preliminary source fingerprints (Table 5). The Mn/V ratios for dust in the study were found to be nearly 10-fold higher compared to previously reported ratios, and this could be due to long term accumulation of Mn emitted from local steel processing industries. However, vehicular exhaust profiles have shown similar Mn/V ratios (0.08–0.53) compared to those reported for Indian vehicular exhaust profiles earlier. In comparison to the reported crustal Cu/Sb ratios, a wide range of Cu/Sb ratios were observed in the current study with lower ratios (indicating high Sb enrichment) for re-suspended dust profiles (1.01–2.31) and higher ratios for vehicular profiles (1.3–60.51). Higher Cu/Sb ratio was observed for 2WVG and 3WVD profiles in comparison to other profiles. V/Ni ratio in PM_{2.5} emissions resulting from vehicular exhaust in the current study were found to be more than five times higher than values reported

elsewhere (Lee *et al.*, 2000; Samara *et al.*, 2003; Moreno *et al.*, 2006; Kong *et al.*, 2011). PRD and UPRD were observed to have similar Ni/V ratios. High Zn/Pb and Zn/Cd ratios in resuspended dust indicate Zn enrichment in the dust, and this could be due to the deposition of exhaust particles, as well as contribution of non-exhaust particles (e.g., tire wear particles) to the road dust. Fe/Al ratio was also analyzed for all the profiles, and the highest ratio was observed for soil. The Fe/Al ratio values for resuspended PM_{2.5} profiles prepared in the study were found to be several times higher compared to those reported earlier for paved roads (25.5) and unpaved roads (71.2) (Patil *et al.*, 2013), and this could be due to iron rich crustal material in the study region, as well the enrichment due to the local Fe processing industries in the region. It is important to note that the current study focused on PM_{2.5} while the reported ratios of Fe/Al are for PM₁₀. Although some differences are expected due to the difference in the size range, crustal species are typically

Table 5. Comparison of elemental ratios for different composite source profiles and other studies.

Source type	Mn/V	Cu/Sb	As/V	V/Ni	Zn/Pb	Zn/Cd	Cu/Zn	Cu/Cd	Cu/Pb
This study									
Soil	56.51	2.31	0.11	0.93	310.81	3418.93	0.15	524.53	47.68
CD	149.46	1.06	0.07	0.78	1537.17	1566.73	0.03	45.69	44.83
PRD	56.42	1.93	5.48	0.33	3.98	95.58	0.04	4.21	0.18
UPRD	19.75	1.01	1.19	0.82	114.22	670.45	0.02	16.16	2.75
2WVG	0.53	60.51	0.03	0.11	10.04	5127.08	0.36	1865.75	3.65
3WVD	0.15	35.16	0.01	0.73	0.67	3171.63	0.1	320.88	0.07
4WVD	0.08	2.02	0.01	0.62	0.16	326.46	0.29	95.5	0.05
HDVD	0.18	1.34	0.03	0.85	0.01	393	1.84	723.5	0.02
Compiled from other studies (International)									
Coal combustion ^a	-	0.5	4.8	0.7	1.9	17	-	-	-
Soil ^{a,b,c}	6.24	0.3–15	0.1–6.0	0.3–8.3	0.4–9.1	8.5–1630	-	-	-
Gasoline+diesel ^{a,d}	-	2.4–63	2.3	0.4	0.02–1.9	0.9–27	-	-	-
Gasoline vehicles ^e	-	315	1.1	0.02	3.4	56	-	-	-
Diesel vehicles ^e	-	700	0.007	0.15	7.6	407	-	-	-
Cement plant ^{b,e}	27.1	7.4	0.03	11	21.9–42	74.5–195	-	-	-
Oil burning ^{e,f}	-	71	0.02	2–4	1.2	190	-	-	-
Road Dust ^b	50.6	-	-	0.6	8.5	200	-	-	-
Construction dust ^b	37.8	-	-	0.57	11.3	68.1	-	-	-
Traffic ^g	-	3-5	-	-	-	-	0.1–1.8	200–600	1.2–3.5
African dust ^h	-	-	-	2.2–8.4	-	-	-	-	-
Compiled from other studies (National)									
Soil (54P25COM) ⁱ	3.74	0.88	0.62	2.46	0.72	50.83	0.35	17.76	0.25
* Construction (43M10UC) ⁱ	15.22	0.33	1.95	0.41	4.10	115.30	0.03	2.89	0.10
Paved road dust (52P25COM) ⁱ	3.08	2.02	0.30	0.62	0.51	12.24	0.29	3.58	0.15
Unpaved road dust (53P25COM) ⁱ	6.53	1.34	0.39	0.85	0.97	5.68	1.84	10.45	1.79
** (Comp-2S2WG-All) ^j	0.28	0.07	0.00	-	7.81	-	0.02	-	0.17
# (Comp-3WD-2) ^j	0.74	0.13	-	3.82	-	-	-	-	0.96
\$ (Comp-LCVD-all) ^j	0.05	1.82	0.08	9.11	108.84	26.65	0.01	0.14	0.59
@ (Comp-HCVD-all) ^j	-	0.33	-	-	6.70	2.77	0.09	0.25	0.61

^a Watson *et al.*, 2001; ^b Kong *et al.*, 2011; ^c Arditoglou and Samara, 2005; ^d Qin *et al.*, 1997; ^e Samara *et al.*, 2003; ^f Lee *et al.*, 2000; ^g Weckwerth, 2001; ^h Moreno *et al.*, 2006; ⁱ CPCB, 2008a; ^j CPCB, 2008b.

* PM₁₀ profile;

** 2 wheeler vehicle-gasoline based (Composite);

3 wheeler vehicle-diesel based (Composite);

\$ 4 wheeler vehicle-diesel based (Composite);

@ Heavy-duty vehicle-diesel based (Composite).

expected to be present in similar concentration in both PM₁₀ and PM_{2.5} fractions.

Relative Source Signatures

To further understand the sources of the different chemical species, relative source signatures were calculated using the following formula (Yang et al., 2002; Kong et al., 2011):

$$\text{Ratio}_{i,j} = \frac{(X_i / \sum X)_j}{(X_i / \sum X)_{\min}} \quad (3)$$

where X_i was the i^{th} individual species concentration, $(X_i / \sum X)_j$ is the quotient of i^{th} individual species ij divided by the sum of 32 species concentrations of emission source j and $(X_i / \sum X)_{\min}$ is the quotient of i^{th} individual species divided by the sum of 32 species concentrations which were the minimum for all emission sources.

Six chemical species with the highest ratio values for total relative source profiles were used as source-specific-species (Table 6) (Mitra et al., 2002; Viana et al., 2008; Watson et al., 2008; Kong et al., 2011). Al, Si, K, Ca and Fe were identified as source markers for dust emissions while OC, EC, S, Pb, and V were identified for vehicular emissions. Na and Zn were identified as markers for the soil profile while Ca, Na and Al were found to be representative species for CD. The source-specific tracers for PRD and UPRD were different compared to other reported species with a prominence of trace metals (Table 6). V, Pb and EC were identified for vehicular exhaust emissions with addition of SO_4^{2-} , NO_3^- and NH_4^+ . The identified source specific species (Table 6) were also cross-compared with marker species. New sets of source signatures for different types of vehicular exhaust and re-suspended dust emissions for Indian origin have been observed compared to those reported elsewhere (Mitra et al., 2002; Viana et al., 2008; Watson et al., 2008).

CONCLUSIONS

Eight dust and vehicular emission source profiles were developed for Raipur city (India) and each source profile included 21 elemental species, 9 water soluble ions, OC and EC. Crustal elements (Al, Ca, Fe, and Mg), OC, and EC were found to be the most abundant species in resuspended dust profiles while Ca, a source marker for construction emissions, was found to be significantly enriched in construction dust. Vehicular emissions were dominated by carbonaceous species (OC and EC) which accounted for 79–92% of the total measured mass. The trace elements were found in higher abundance in vehicular emission profiles compared to resuspended dust profiles. Mass reconstruction was attempted and good results were obtained with respect to the measured mass. Markers identified for emission sources include Al, Ca, Zn, Mg^{2+} and SO_4^{2-} for resuspended dust profiles and EC, V and Pb with some ionic species for vehicular emission. This is the first attempt at detailed chemical characterization of traffic and dust profiles in the Raipur region, and one of the few studies to characterize PM_{2.5} profiles for India. Results highlight the need of more

Table 6. Source signature for vehicular and resuspended dust profiles.

Source type	Source signature	Source type	Source signature
	Geological sources		Vehicular sources
Soil	Na^+ , SO_4^{2-} , Zn, Se, K^+ , Cl^-	2WVG	F^- , Cr, Cd, V, Na^+ , Ni
CD	Zn, Na, Mo, Al, Mg^{2+} , Ca	3WVD	Ca^{2+} , Mg^{2+} , NH_4^+ , K, Se, SO_4^{2-}
PRD	Na^+ , SO_4^{2-} , As, F^- , Mg^{2+} , Se	4WVD	F^- , NO_3^- , Cd, Pb, SO_4^{2-} , EC
UPRD	Na^+ , SO_4^{2-} , F^- , Mg^{2+} , Se, Pb	HDVD	F^- , NH_4^+ , Se, Pb, SO_4^{2-} , EC
City dust ^f	Al, Si, Ca, Fe, K	On-road vehicle ^{a,b,c}	OC, EC, NH_3 , S, Fe, Zn, Cu, Pb, Ba, Br
Road dust(paved and unpaved) ^a	Al, Si, K, Ca, Fe	Construction dust ^a	Al, Si, K, Ca, Fe

^aWatson et al., 2008;

^bMitra et al., 2002;

^cVianna et al., 2008.

detailed locally developed PM_{2.5} source profiles (inclusions of Si, Ti, thermally fractionate OC and EC, molecular organic markers) for different regions of the country. Results from the study can be used in receptor modeling studies (in case of CMB, PMF, UNMIX) as well as in emission inventory calculations.

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SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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