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PM_{2.5} Chemical Source Profiles of Emissions Resulting from Industrial and Domestic Burning Activities in India

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ABSTRACT

A study has been performed to develop PM_{2.5} (particles with aerodynamic diameters ≤ 2.5) chemically speciated source profiles of different industrial and domestic burning practices in India. A total of fifty-five PM_{2.5} samples have been collected in emissions resulting from (1) industrial furnaces, (2) household fuels, (3) municipal solid waste burning, and (4) welding workshop burning practices, and categorized for eleven subtypes of sources. The collected samples were subjected to chemical analysis for twenty-one elemental (Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, Pb, S, Sb, Se, V, Zn), nine ionic (Na⁺, K⁺, Mg²⁺, Ca²⁺, NH₄⁺, Cl⁻, F⁻, NO₃⁻, SO₄²⁻), OC, and EC source indicator species using atomic absorption spectrometry, ion chromatography and carbon analysis (thermal/optical transmittance method), respectively. The carbonaceous fraction was most abundant in household fuel burning emissions ($47.6 \pm 7.45\%$ to $65.92 \pm 13.13\%$). The ionic/elemental ratios of major inorganic constituents (Ca²⁺/Ca, Mg²⁺/Mg and Na⁺/Na) have been identified to describe the PM_{2.5} emissions from combustion or re-suspension dusts during industrial activities. Brick Kiln processes (BKP) have been identified as the major emitter of the highest number of toxic species (Cd, Co, Mo, Sb and V), followed by steel re-rolling mills (Hg and Pb) and steel processing industries (As, Ni). The source marker calculations also confirmed that K⁺, Mn, and As are good markers for biomass burning, metallurgical industrial emission, and coal burning, respectively, similar to the findings in previous studies.

Keywords: Source profiles; PM_{2.5}; Source markers; Industrial source; Household fuel burning emission.

INTRODUCTION

The source apportionment of PM fractions increases with different trends using receptor models, mostly chemical mass balance (CMB), to develop pollution control and mitigation strategies worldwide (Watson *et al.*, 2002; Samara, 2005; Khan *et al.*, 2010; Kong *et al.*, 2010). Receptor models that derive profiles from ambient/indoor measurements require systematic chemically speciated emission profiles from prominent sources that were possible to effect pollutant concentration at receptor for verification (Hopke, 1999; Watson *et al.*, 2001, 2002; Brook *et al.*, 2003; Gupta *et al.*, 2007). These source profiles are the fractional mass (abundances \pm uncertainty) of measured chemical species relative to primary PM mass of source emissions (Watson *et*

al., 2001) and one of the most important parameters (Pant and Harrison, 2012) to: 1) create chemically speciated emission inventories (Cass and McRae, 1983; Kuykendal *et al.*, 1990; Chow *et al.*, 2004), 2) apportion receptor concentrations to source (Watson *et al.*, 1984, 1990, 1991, 2001) and 3) estimate toxic and hazardous pollutant emissions (Chow *et al.*, 2004). Chemical abundance in most of earlier source profiles is accompanied by an uncertainty/standard deviation value that intends to represent the errors/variability of that abundance resulting from differences among separate emitters and between samples taken same/different times from the same emitters; which is essential to CMB runs (Watson *et al.*, 1994, 2001; Chow *et al.*, 2003; Ho *et al.*, 2003; Chow *et al.*, 2004; Tsai *et al.*, 2007).

Several source profiles of different PM fractions have been developed for different individual sources and applied widespread (Chow and Watson, 1994; Vega *et al.*, 2001; Watson and Chow, 2001; Watson *et al.*, 2001; USEPA, 2002; Chow *et al.*, 2004; Yatkin and Bayram, 2008). These profiles differ with sources, process operating conditions, geology, and geographic seasonality (Watson *et al.*, 2001; Kong *et al.*, 2011; Pant and Harrison, 2012). Additional profiles are

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always needed for contemporary inventories and source apportionment studies (Watson *et al.*, 2001). Very limited source emission chemical profiles of PM_{2.5} have been developed in Indian context (Gadkari and Pervez, 2007, 2008; CPCB, 2008; Patil *et al.*, 2013).

The current study presented PM_{2.5} chemical source profiles of emissions resulting from 07 different industrial processes including arc-welding workshops and 04 domestic burning practices involved with household cooking activities and municipal solid waste management in India. These PM_{2.5} chemical source profiles were developed with the objectives to meet the requirement of location specific and latest source profiles that could be applied for chemical mass balance receptor modelling studies; and to update previous source profiles.

METHODOLOGY

The development of PM_{2.5} chemical source profiles of selected burning practices have been carried out as a part of a comprehensive source apportionment study of indoor/outdoor PM_{2.5} in a dense urban- industrial zone of India (Balakrishna and Pervez, 2009; Pervez *et al.*, 2012) by following a real-world pooled sampling plan using purposeful

study design (Gilbert, 1987). Sampling of PM_{2.5} was conducted at four different types of combustion sources, mainly observed in urban areas of Chhattisgarh, India: (1) Municipal waste burning, (2) Household fuel burning (3 sub-types), (3) Mineral based coal fired industries (6 sub-types) and (4) Fabrication workshops (Table 1) (Balakrishna and Pervez, 2009, 2011; Pervez *et al.*, 2012).

Description of Study Area and Source Characteristics

Raipur-Bhilai, major industrial cities of Chhattisgarh, India located in global scale of 21°14'22.7"N, 81°38.1"E and 21°11'0"N, 81°23'6"E respectively, having population 1,635,784 (Census, 2011), is known for most dense heavy industrial zone composed of mainly iron processing, thermal power generation and cement production activities. Identification of PM_{2.5} emission sources were based on previous reported air monitoring studies (Dubey and Pervez, 2008; Balakrishna *et al.*, 2011; Pervez *et al.*, 2012), layout map, and a survey of current burning practices involved with industrial and domestic activities. About 1200 tonnes of municipal solid waste (MSW) is generated every day in Raipur-Bhilai, region and about 65% of the MSW (about 650 tonne) was disposed-off using open burning procedure on daily basis. Pervez *et al.* (2012) reported that all populations

Table 1. Description of domestic and industrial source characterization, material used in burning practices and sampling method.

S.N.	Profile Code	Source Name	Source frequency	Type & Material burnt	Adopted sampling methodology
Municipal solid waste burning source					
1.	MSWB	Municipal solid waste burning	Over than ~5 major dumping zone and over than ~200 minor burning places	Synthetic and natural biomaterials in a 1:8 ratio	In-plume
Household fuel burning sources					
1.	RSFS	Residential solid fuel stoves	45% households	Conventional mixture of Coal, wood, dung with ratio in 2:1:1	In-plume
2.	RKS	Residential kerosene stoves	30% households	Kerosene	In-plume
3.	RLPGS	Residential LPG stoves	25% households	Liquid petroleum gas	In-plume
Mineral based coal fired industries					
1.	CPI	Cement production industry	> 9 major and > 12 minor units	Limestone, Gypsum, Steel slag	Chamber re-suspension of bag-filter dust
2.	FEMNI	Ferroy-Manganese industry	> 90 major and minor units	Iron ore, Coal, Dolomite	Chamber re-suspension of bag-filter dust
3.	SPI	Steel production industry	> 130 major and minor units	Iron ore, Coal, Dolomite	Chamber re-suspension of bag-filter dust
4.	CTPP	Coal based thermal power plant	> 50 major and minor units	Coal	Chamber re-suspension of bag-filter dust
5.	SRM	Steel rolling mills	> 150 major and minor units	Scrap cuttings, Steel ingots, Coal	Chamber re-suspension of bag-filter dust
6.	BKP	Brick kiln process	> 300 major and minor furnaces	Brick clay, Coal, Wood	In-plume
Workshops					
1.	EAW	Fabrication and welding	> 1200 major and minor units	Iron, welding material	In-plume

of this area use three categorized stoves (based on fuels) for household burning purposes, namely- Liquid petroleum gas (LPG) stoves, Kerosene stoves and stoves with conventional solid fuels. The census of India (2011) describes the household statistical figures of different type of fuel use for cooking purposes: 25% LPG stoves, 30% kerosene stoves and 45% stoves with conventional solid fuel in Central India. As far as different industrial processes carrying out in the study region are concern, about 1351 industries are currently existing in the study region; out of that heavy, medium and small scale industries numbers are 114, 295 and 942, respectively (DoCI, 2012). Nearly 300 major and medium units of iron processing with consumption of 20 Million Tonne (MT) of iron ore/scrap steels per year, ~50 small, medium and major units of coal-burning power generation with coal consumption of 19.03 MT/yr, 15 units of cement production with lime stones/slag/gypsum consumption of 26 MT/yr occurred in previous years. A total of 32.11 MT/yr coals are consumed in production of power generation, steel processes and cement production in the study area. Accordingly, eleven different types of combustion/burning practices involved with industrial, household and outdoor activities have been chosen for the development of PM_{2.5} chemical source profiles. Details of source types, justification of their selections, combustion material used have been described in Table 1 and Fig. 1.

Sampling

Two different sampling methodologies have been adopted according to nature and characteristics of different burning practices: real-world in-plume and re-suspension sampling

(Chow *et al.*, 2004; Patil *et al.*, 2013). In case of industrial combustion processes, stack emitted bag filter house dust samples were re-suspended to collect PM_{2.5} fractions using standard procedures reported elsewhere (Chow *et al.*, 2004; Gadkari and Pervez, 2007). In case of open burning sources related to household fuel burning (RSFS, RKS, RLPGS), outdoor municipal waste burning practices (MSWB), arc-welding workshops (EAW) and brick kilns (BKP), PM_{2.5} was sampled from smoke plume. All these open burning sources does not have stacks and PM_{2.5} impactors were positioned in smoke plume. Before in-plume sampling, background PM_{2.5} were measured for subtraction from the in-plume concentration (Chakrabarty *et al.*, 2013; Dewangan *et al.*, 2013). In case of PM_{2.5} sampling using chamber re-suspension procedures, sampling duration was optimized according to standard filter loading conditions (Chow *et al.*, 2003; DRI, 2011). In case of in-plume sampling for open burning sources, PM_{2.5} sampling event was conducted in 3–4 episodes to cover whole burning processes (Chow *et al.*, 2003; DRI, 2011; Chakrabarty *et al.*, 2013). PM_{2.5} has been collected on quartz fiber filters (QFF) (1851-047, Whatman, UK) using Parallel operation of five PM_{2.5} samplers (MINIVOL, Ver. 4.2, Model AirMetrics) in each source site at average flow rate of 5 L/min. Filter selection, preparation, calibration, installation, transportation, preservation, weighing measurements and field blanks were conducted by following the quality control and quality assurance described in air sampling protocol reported elsewhere (USEPA, 1999a, b; CPCB 2008; DRI, 2011; Patil *et al.*, 2013). Details of PM_{2.5} source sites, their location frequencies in study region and sampling methodology used has been described in Table 1.

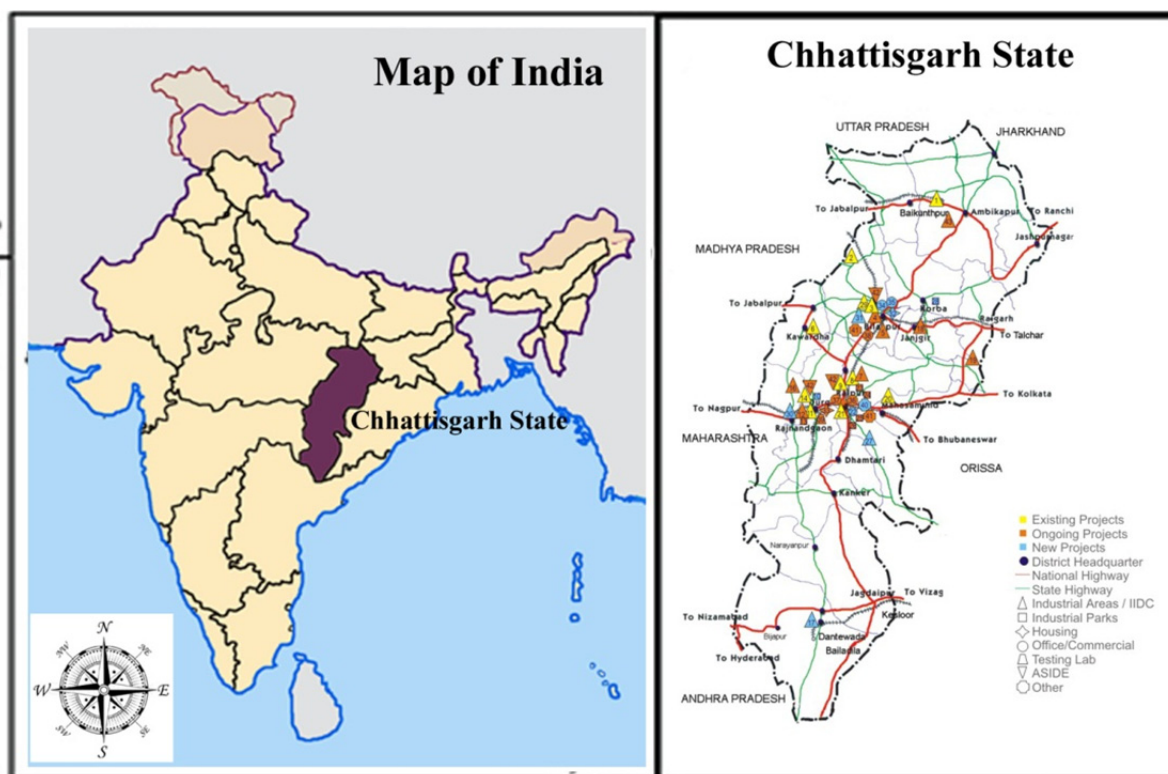


Fig. 1. Location map of study region, an urban-industrial environment, Raipur, India.

Chemical Analysis

32 Chemical species [Twenty one elemental- Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Mo, Na, Ni, Pb, S, Sb, Se, V, Zn; nine water soluble ions- Na^+ , K^+ , Mg^{2+} , Ca^{2+} , NH_4^+ , Cl^- , F^- , NO_3^- , SO_4^{2-} and carbonaceous fractions- Organic (OC) and Elemental (EC) carbon], known for source marker species have been determined in $\text{PM}_{2.5}$. In case of inorganic constituents, water extractable (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , Cl^- , F^- , NO_3^- and SO_4^{2-}) and acid digested (1:3 $\text{H}_2\text{O}_2/\text{HNO}_3$) (Na, Mg, Al, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Pb, Mo, Se, Sb and Hg) were quantified separately (Katz, 1977; Chow et al., 2003). Elemental species (Al, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Pb, Mo, Se, Sb and Hg) and selected water extractable (Na, Mg, K and Ca) were determined graphite furnace based atomic absorption spectrophotometrically (AAS) and cold vapour-AAS (iCE3500 Model, Thermo Fisher) using recommended conditions of operation and reported protocol of analysis (Thermo Fisher, 2008; DRI, 2011). Anions in water extracts (F^- , Cl^- , NO_3^- , SO_4^{2-}) and digested samples (SO_4^{2-}) were determined ion chromatographically (2000 Model, Dionex) using reported protocol of analysis (Chriswell et al., 1986; Watson et al., 1999; Dionex, 2005; Kulshrestha et al., 2010). SO_4^{2-} measurements in both water extracts and digested samples of $\text{PM}_{2.5}$ has been carried out to quantify sulphur content of $\text{PM}_{2.5}$ by subtracting the SO_4^{2-} (digested) from SO_4^{2-} (water extracts) (Gurugubelli et al., 2013). NH_4^+ , a marker of biomass burning, is determined spectrophotometrically (1305E Model, Systronics) using recommended procedures (Harrison and Perry, 1986).

As far as OC and EC analysis is concern, thermal optical transmittance (TOT) method presented by National Institute for Occupational Safety and Health- NIOSH-5040 protocol (NIOSH, 1999) using semi-continuous thermal/optical carbon analyzer (Sunset Laboratory, Model 4L, USA) (Birch and Cary, 1996; Schauer, et al., 2003; Pipal et al., 2014) has been adopted. Concentration of selected chemical species measured in $\text{PM}_{2.5}$ were corrected by subtracting them from those found in field blanks; followed by subtraction from those found in background $\text{PM}_{2.5}$. The background corrected concentrations along with their uncertainties and in-plume/background ratio (for those measured in smoke plume) have been presented in Table 2 and Table 3. Uncertainty of source profile abundances estimated as the standard deviation of the average from five source tests (Watson et al., 2001; Watson and Chow, 2007). Species concentrations in laboratory blank and field blanks have also been presented in Table S1 (supporting information). Chemical species abundances in four different ranges of percent by weight along with comparison with reported values have been presented in Table 4 and Table 5. Components of crustal origin, ionics, trace elements and carbonaceous matter found in selected source profiles have been presented in Fig. 2. Source markers of selected source emitted $\text{PM}_{2.5}$ has been shown in Table 6.

RESULTS AND DISCUSSION

Description of $\text{PM}_{2.5}$ Chemical Source Profiles

A source profile comprised offivefractional abundances

of individual chemical species with respect to total PM mass and individual uncertainty or standard deviation value for particular chemical species (Watson et al., 2001). Chemical profiles have shown discernible pattern in relative strengths of selected species in $\text{PM}_{2.5}$ emissions resulting from industrial and domestic burning practices. Profile-wise measured mass accounted were: 49.30 ± 3.35% (MSWB), 62.64 ± 5.26% (RSFS), 78.05 ± 5.07% (RKS), RLPGS (74.03 ± 3.85%), CPI (50.65 ± 3.60%), FEMNI (46.18 ± 4.02%), SPI (43.04 ± 3.57%), CTPP (33.44 ± 1.71%), SRM (58.33 ± 3.62%), EAW (57.78 ± 3.93%), and BKP (60.31 ± 4.89%); unaccounted content might be due to oxides and water content (Ho et al., 2003; Watson et al., 2012) and silica which were not measured directly by the methods applied to the source emissions. On analysing the association of different chemical components (OC/EC, crustal origin, ionics and trace elements) with $\text{PM}_{2.5}$ emission source profiles (Fig. 2), significant variation in OC and EC content of $\text{PM}_{2.5}$ with variability of 73.75% and 96.88%, respectively, across the selected source sites is observed; similar to earlier reported source profiles (Watson et al., 2001; Chow et al., 2003, 2004; Kong et al., 2011). Total carbonaceous matter (TC) (sum of OC and EC) is accounted for > 50% of $\text{PM}_{2.5}$ emissions in most of the source sites with OC/EC ratio > 1; attributed to incomplete combustion activities resulting in higher emissions of organic carbon in smouldering phases (Chakrabarty et al., 2013). Species of crustal origin (Al, Mn, Mg, Ca, Fe, K and Na) were also found in different proportions across the source profiles with highest variability of 117.42%; comparable with those reported earlier (Chow et al., 2003; Kong et al., 2011). As far as trace elements (Cu, Zn, As, Pb, Cr, Ni, Co, Cd, Hg, V, Mo, Sb, Se and S) and water soluble ions (Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , F^- , Cl^- , NO_3^- , SO_4^{2-}) are concern, more similarity (65.47% and 35.38%, respectively) compared to carbonaceous fractions and crustal species has been observed (Samara et al., 2003; Chow et al., 2004; Patil et al., 2013). Detailed $\text{PM}_{2.5}$ chemical profiles have been presented in Table 2 and Table 3.

Municipal Solid Waste Burning (MSWB)

Municipal refuse open burning is one of the major contributors to the air pollution in Indian cities (Patil et al., 2013). $\text{PM}_{2.5}$ emissions from MSWB is prominently loaded with TC (84% of measured mass) having OC/TC ratio of 0.94 and potassium (5.14% of measured mass) having K^+/K ratio of 0.77; comparable to those reported for vegetative and biomass burning (Watson et al., 1994, 2001; Chow et al., 2004). Abundance of other ions (Na^+ , NH_4^+ , Ca^{2+}) were significantly higher (> 1% of measured mass) than species related to crustal origin and trace elements.

Residential Fuel Stoves

Residential cooking stoves are mostly uses three different types of fuel/fuel mixtures for cooking purposes in India: solid fuel mixture (3:1:1 ratio of Coal, wood, dung cakes), kerosene and LPG. Highest abundance of TC (75–87% of measured mass) in $\text{PM}_{2.5}$ emissions has been observed. In addition, RSFS has shown OC/EC ratio of 6.9; two and

Table 2. PM_{2.5} composite sources profiles (weight percent by mass) of emissions from residential fuel and municipal waste burning practices.

Species	MSWB	In-plume/ Background	RSFS	In-plume/ Background	RKS	In-plume/ Background	RLPGS	In-plume/ Background
Al	0.03608 ± 0.01201	0.00	0.02639 ± 0.01510	0.00	0.46045 ± 0.16270	0.05	0.39362 ± 0.08894	0.04
As	0.00083 ± 0.00020	0.33	0.05400 ± 0.01000	21.72	0.16500 ± 0.04310	66.38	0.00020 ± 0.00021	0.08
Ca	0.60241 ± 0.06713	0.34	0.76957 ± 0.25100	0.43	0.01645 ± 0.00624	0.01	0.01432 ± 0.01008	0.01
Cd	0.00456 ± 0.00215	2.13	0.00004 ± 0.00010	0.02	0.08000 ± 0.01000	37.46	0.00450 ± 0.00510	2.11
Co	0.86000 ± 0.02100	173.63	0.02500 ± 0.00200	5.05	0.01600 ± 0.00900	3.23	0.00500 ± 0.00300	1.01
Cr	0.08790 ± 0.03100	0.37	0.72000 ± 0.03500	3.01	0.01189 ± 0.01130	0.05	0.06846 ± 0.07500	0.29
Cu	0.06812 ± 0.01615	0.09	0.04583 ± 0.01330	0.06	0.00390 ± 0.00140	0.01	0.00670 ± 0.00180	0.01
Fe	0.28002 ± 0.03179	1.01	0.15876 ± 0.39370	0.57	0.11200 ± 0.01386	0.40	0.02264 ± 0.03261	0.08
Hg	0.00351 ± 0.00190	0.11	0.00176 ± 0.00300	0.06	0.02620 ± 0.00291	0.83	0.00762 ± 0.00914	0.24
K	2.51720 ± 0.63300	2.55	4.71310 ± .52314	4.78	1.20110 ± 0.49600	1.22	0.39500 ± 0.13114	0.40
Mg	0.07429 ± 0.05845	0.08	0.17730 ± 0.06000	0.19	0.00133 ± 0.00963	0.00	0.00409 ± 0.00356	0.00
Mn	0.04575 ± 0.01057	0.04	0.02198 ± 0.01670	0.02	0.00230 ± 0.00161	0.00	0.02927 ± 0.00819	0.02
Mo	0.00228 ± 0.00027	4.76	0.00165 ± 0.00660	3.45	0.00861 ± 0.00215	17.98	0.07470 ± 0.00561	156.04
Na	0.77178 ± 0.45500	0.57	0.88587 ± 1.23000	0.66	0.06136 ± 0.04561	0.05	0.97956 ± 0.86112	0.73
Ni	0.01147 ± 0.00185	0.11	0.01454 ± 0.05200	0.13	0.56510 ± 0.02881	5.21	0.02777 ± 0.01214	0.26
Pb	0.00120 ± 0.00025	0.00	0.00002 ± 0.00005	0.00	0.92000 ± 0.01300	2.81	0.00132 ± 0.00052	0.00
S	0.01062 ± 0.00300	0.00	0.10234 ± 0.02601	0.05	1.71874 ± 0.26299	0.76	0.69779 ± 0.16968	0.31
Sb	0.00015 ± 0.00007	0.04	0.00015 ± 0.00030	0.04	0.28000 ± 0.11400	75.28	0.04400 ± 0.02100	11.83
Se	0.00839 ± 0.00095	11.11	0.00215 ± 0.00025	2.85	0.62500 ± 0.04900	827.90	0.05300 ± 0.00650	70.21
V	0.00837 ± 0.00860	3.44	0.00318 ± 0.00061	1.31	0.65030 ± 0.08500	267.56	0.01052 ± 0.00378	4.33
Zn	0.30428 ± 0.02434	0.50	0.14362 ± 0.05310	0.24	0.96380 ± 0.01561	1.59	0.03557 ± 0.00279	0.06
F ⁻	0.04000 ± 0.05000	0.13	0.50000 ± 0.02000	1.65	0.17450 ± 0.08921	0.58	0.00012 ± 0.00009	0.00
Cl ⁻	0.41299 ± 0.15743	0.15	1.70187 ± 0.11260	0.60	1.08300 ± 0.03000	0.38	0.64548 ± 0.28828	0.23
NO ₃ ⁻	0.24600 ± 0.08500	0.05	3.48019 ± 0.27100	0.75	1.03600 ± 0.03040	0.22	0.62117 ± 0.01441	0.13
SO ₄ ²⁻	0.49773 ± 0.19170	0.07	1.21811 ± 0.40964	0.18	4.14460 ± 0.22483	0.62	1.77885 ± 0.34721	0.27
Na ⁺	0.74193 ± 0.41125	0.74	0.39359 ± 0.47200	0.39	0.02150 ± 0.00900	0.02	0.79632 ± 0.24871	0.80
NH ₄ ⁺	0.81130 ± 0.02120	0.28	0.27019 ± 0.04920	0.09	4.34420 ± 0.02570	1.48	2.19500 ± 0.03900	0.75
K ⁺	1.92720 ± 0.21210	3.44	4.57750 ± 0.36000	8.16	1.15500 ± 0.20360	2.06	0.35851 ± 0.02700	0.64
Ca ²⁺	0.47695 ± 0.20500	0.76	0.71947 ± 0.05100	1.15	0.00946 ± 0.00400	0.02	0.01283 ± 0.01012	0.02
Mg ²⁺	0.05400 ± 0.00170	0.19	0.13000 ± 0.01500	0.46	0.00110 ± 0.00090	0.00	0.00400 ± 0.00313	0.01
OC	39.07940 ± 6.87439	1.84	41.61166 ± 6.74153	1.95	45.19647 ± 3.18710	2.12	37.91570 ± 12.83300	1.78
EC	2.50984 ± 0.95920	0.64	5.98930 ± 0.70900	1.53	14.17995 ± 2.06280	3.61	28.00000 ± 0.30000	7.13
TC	41.58924 ± 7.83359		47.60096 ± 7.45053		59.37642 ± 5.24990		65.91570 ± 13.13300	
Sum%	49.29647 ± 3.35215		62.63857 ± 5.26163		78.04825 ± 5.07313		74.03195 ± 3.84966	

* Ca²⁺, K⁺, Na⁺, Mg²⁺ and TC were eliminated for double counting in sum%.

Table 3. PM_{2.5} composite sources profiles (weight percent by mass) of emissions from industrial burning practices.

Species	CPI	FEMNI	SPI	CTPP	SRM	EAW	In-plume/ Background	BKP	In-plume/ Background
Al	3.79199 ± 0.30528	2.46541 ± 0.30952	0.77095 ± 0.10408	5.24498 ± 0.60980	0.08400 ± 0.00598	2.95246 ± 0.19963	0.33	0.09032 ± 0.00301	0.01
As	0.00010 ± 0.00013	0.00710 ± 0.00210	0.05500 ± 0.03000	0.27300 ± 0.07300	0.00393 ± 0.00093	0.10581 ± 0.00948	42.57	0.10733 ± 0.03145	43.18
Ca	20.0980 ± 0.25756	6.54080 ± 1.27759	1.12100 ± 0.39350	1.62692 ± 0.78370	5.5807 ± 0.39318	1.39425 ± 0.10591	0.78	2.03986 ± 0.94582	1.13
Cd	0.00028 ± 0.00127	0.00019 ± 0.00015	0.00052 ± 0.00032	0.00005 ± 0.00009	0.00238 ± 0.00020	0.01352 ± 0.00137	6.33	0.05696 ± 0.00603	26.67
Co	0.00202 ± 0.00100	0.00182 ± 0.00194	0.00055 ± 0.00015	0.00273 ± 0.00050	0.00036 ± 0.00021	0.00258 ± 0.00187	0.52	0.00901 ± 0.00634	1.82
Cr	0.03372 ± 0.00305	0.71700 ± 0.06000	0.96280 ± 0.01600	0.09077 ± 0.02470	0.00027 ± 0.00008	0.06146 ± 0.00605	0.26	0.01489 ± 0.00305	0.06
Cu	0.94700 ± 0.01000	0.82500 ± 0.00600	0.67500 ± 0.01800	0.11047 ± 0.08000	0.00782 ± 0.00061	0.15264 ± 0.00922	0.21	0.09042 ± 0.00926	0.12
Fe	3.03099 ± 0.24105	6.81153 ± 3.79153	15.27590 ± 0.32431	1.81559 ± 0.43860	5.08803 ± 0.35994	31.97249 ± 2.26139	115.17	0.44480 ± 0.07704	1.60
Hg	0.00888 ± 0.00381	0.00913 ± 0.01544	0.00785 ± 0.00340	0.00544 ± 0.00040	0.01001 ± 0.00085	0.07021 ± 0.00610	2.23	0.00874 ± 0.00514	0.28
K	0.61121 ± 0.02858	0.54332 ± 0.12590	1.09798 ± 0.35200	1.12695 ± 0.13240	0.71173 ± 0.05723	6.60688 ± 0.53671	6.70	0.42686 ± 0.43935	0.43
Mg	5.84425 ± 0.53912	1.95458 ± 0.42539	0.99930 ± 0.15090	0.92400 ± 0.16965	1.08633 ± 0.07691	1.96525 ± 0.18215	2.10	0.38074 ± 0.25303	0.41
Mn	0.05213 ± 0.00699	1.97486 ± 0.08710	0.20133 ± 0.02443	0.71051 ± 0.06500	0.05624 ± 0.00400	5.04121 ± 0.30103	4.11	0.01540 ± 0.00189	0.01
Mo	0.00165 ± 0.00022	0.01261 ± 0.00363	0.00489 ± 0.00049	0.00004 ± 0.00001	0.00032 ± 0.00023	0.00656 ± 0.00186	13.70	0.02089 ± 0.00961	43.64
Na	3.26488 ± 1.13284	3.39504 ± 0.47439	0.67693 ± 0.39821	0.57435 ± 0.08027	1.34385 ± 0.09567	1.09229 ± 0.20063	0.81	0.96783 ± 0.10926	0.72
Ni	0.00333 ± 0.01905	0.02519 ± 0.00703	0.04276 ± 0.00111	0.02160 ± 0.00172	0.00654 ± 0.00168	0.00998 ± 0.00995	0.09	0.03296 ± 0.04081	0.30
Pb	0.02974 ± 0.00267	0.00341 ± 0.00072	0.00461 ± 0.00051	0.04959 ± 0.00400	2.22579 ± 0.15830	0.60203 ± 0.05303	1.84	0.02857 ± 0.00560	0.09
S	0.98579 ± 0.04756	0.02215 ± 0.00135	0.00735 ± 0.00014	0.87193 ± 0.02094	0.18249 ± 0.01257	0.00425 ± 0.00215	0.00	0.06215 ± 0.00158	0.03
Sb	0.00196 ± 0.00028	0.00095 ± 0.00013	0.00035 ± 0.00012	0.00014 ± 0.00200	0.0103 ± 0.00074	0.00705 ± 0.00643	1.89	0.03926 ± 0.03095	10.56
Se	0.00130 ± 0.00038	0.00434 ± 0.00251	0.00158 ± 0.00080	0.05200 ± 0.00400	0.00095 ± 0.00068	0.02816 ± 0.00684	37.30	0.02083 ± 0.01079	27.59

Table 3. (continued).

Species	CPI	FEMNI	SPI	CTPP	SRM	EAW	In-plume/ Background	BKP	In-plume/ Background
V	0.00187 ± 0.00394	0.01002 ± 0.00704	0.02653 ± 0.00496	0.01854 ± 0.00211	0.00670 ± 0.00078	0.01951 ± 0.00489	8.03	0.08092 ± 0.02066	33.29
Zn	0.31990 ± 0.03213	0.73586 ± 0.20121	0.46236 ± 0.05455	0.15036 ± 0.01353	0.78549 ± 0.05557	2.10060 ± 0.07025	3.46	0.08909 ± 0.00813	0.15
F ⁻	0.23748 ± 0.08865	0.17410 ± 0.05500	0.71000 ± 0.02300	1.73480 ± 0.16500	0.01366 ± 0.00116	0.40982 ± 0.03096	1.36	0.07190 ± 0.03106	0.24
Cl ⁻	0.01654 ± 0.01059	0.47000 ± 0.17425	1.13534 ± 0.03970	0.21566 ± 0.07950	0.90901 ± 0.07097	0.27349 ± 0.05170	0.10	0.60190 ± 0.04750	0.21
NO ₃ ⁻	0.00299 ± 0.00175	0.45055 ± 0.31774	0.08527 ± 0.00621	0.40000 ± 0.20000	0.39439 ± 0.02933	0.01189 ± 0.00854	0.00	0.70456 ± 0.02140	0.15
SO ₄ ²⁻	3.31847 ± 0.77165	0.73854 ± 0.00447	0.53500 ± 0.00012	1.16403 ± 0.06695	4.17298 ± 0.29597	0.51454 ± 0.04012	0.08	0.83246 ± 0.00589	0.13
Na ⁺	1.06891 ± 0.36055	1.98625 ± 0.17927	0.41815 ± 0.12630	0.39449 ± 0.03665	0.24446 ± .02976	0.81442 ± 0.06250	0.82	0.78128 ± 0.56128	0.78
NH ₄ ⁺	0.00956 ± 0.00314	1.11496 ± 0.41855	0.05175 ± 0.01115	0.38322 ± 0.11700	0.13700 ± 0.01484	0.31450 ± 0.08500	0.11	0.62025 ± 0.30722	0.21
K ⁺	0.01276 ± 0.01140	0.24413 ± 0.12670	0.70123 ± 0.08280	0.82600 ± 0.05030	0.05234 ± 0.01421	3.31681 ± 0.25985	5.91	0.26115 ± 0.17726	0.47
Ca ²⁺	2.55696 ± 0.19926	0.76072 ± 0.17605	0.80390 ± 0.01170	1.13420 ± 0.02670	2.33327 ± 0.08260	0.64688 ± 0.20334	1.03	1.41742 ± 0.70371	2.26
Mg ²⁺	0.00236 ± 0.00081	0.49190 ± 0.01306	0.55240 ± 0.01420	0.67000 ± 0.02500	0.34232 ± 0.03474	0.29062 ± 0.06212	1.03	0.11199 ± 0.01093	0.40
OC	7.89808 ± 1.59461	11.59053 ± 0.65233	4.37481 ± 0.45575	9.16596 ± 0.19555	24.06154 ± 1.20308	1.88401 ± 0.09420	0.09	20.53330 ± 0.37666	0.96
EC	0.14000 ± 0.05097	5.58571 ± 0.43274	13.74898 ± 1.82446	6.70770 ± 0.32630	11.47508 ± 0.57375	0.16661 ± 0.00833	0.04	31.92240 ± 1.59610	8.13
TC	8.03808 ± 1.64558	17.17624 ± 1.08507	18.12379 ± 2.28021	15.87366 ± 0.52185	35.53662 ± 1.77683	2.05062 ± 0.10253		52.45570 ± 1.97276	
Sum%	50.65410 ± 3.59664	46.18473 ± 4.01807	43.03669 ± 3.57204	33.44134 ± 1.70550	58.32601 ± 3.61621	57.78406 ± 3.92908		60.31460 ± 4.88548	

* Ca²⁺, K⁺, Na⁺, Mg²⁺ and TC were eliminated for double counting in sum%.

Table 4. Summary of chemical species abundance in PM_{2.5} emissions from domestic burning practices and their comparison with nationally and internationally reported profiles.

Source ID	Various studies	Chemical abundances in percent mass		
		<0.1%	0.1–1%	1–10%
MSWB	Present study*	Al, As, Cd, Cr, Cu, Hg, Mg, Mn, Mo, Na, Ni, Pb, S, Sb, Se, V, F⁻, Mg²⁺	Ca, Co, Fe, K, Zn, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, Ca²⁺	K⁺, EC
	<i>USEPA Speciate 4.0</i>	<i>Sb, As, Cd, Ce, Cr, Cu, Mn, Mo, Ni, Pd, Se, Sn</i>	<i>Al, NH₄⁺, Ca, EC, Fe, Pb, Hg, OC, K, Na, SO₄²⁻, S</i>	<i>OC</i>
	<i>CPCB, 2008***</i>	<i>Al, Mg, As, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, V, Mg²⁺</i>	<i>Ca, Fe, SO₄²⁻, Ca²⁺, EC</i>	<i>OC</i>
RSFS	Present study	Al, As, Cd, Co, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, V	Ca, Cr, Fe, Mg, Na, S, Zn, F, Na, NH₄⁺, Ca²⁺, Mg²⁺	K, Cl⁻, NO₃⁻, SO₄²⁻, K⁺, EC, OC
	<i>USEPA Speciate 3.2</i>	<i>Al, As, Ca, Cl, Co, Cu, Fe, Pb, Mg, Mn, Hg, Mo, Na, S, Zn</i>	<i>NH₄⁺, Cl⁻, NO₃⁻, K, K⁺, SO₄²⁻</i>	<i>EC, OC</i>
	<i>PR.No.-423312.5</i>	<i>Al, Ca, Fe, Mg, As, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, V, NO₃⁻, NH₄⁺, Mg²⁺</i>	<i>K, Cl⁻, K⁺, EC</i>	<i>OC</i>
RKS	Present study	Ca, Cd, Co, Cr, Cu, Hg, Mg, Mn, Mo, Na, Ni, Pb, Sb, Se, V, NO₃⁻, NH₄⁺, Mg²⁺	Al, As, Fe, Ni, Pb, Sb, Se, V, Zn, F⁻, NH₄⁺, K⁺	OC, EC
RLPGS	Present study	As, Ca, Cd, Co, Cr, Cu, Fe, Hg, Mg, Mn, Mo, Ni, Pb, Sb, Se, V, Zn, F⁻, Ca²⁺, Mg²⁺	Al, K, Na, S, Cl⁻, NO₃⁻, Na⁺, K⁺	OC, EC
	<i>USEPA Speciate 4.3</i>	<i>Cr, Fe, Mn, Ni, Se, V</i>	<i>Ca, Na, S, Cl⁻, NO₃⁻, EC</i>	<i>SO₄²⁻, OC</i>
	<i>PR. No.-91156</i>	<i>Al, Ca, Mg, Cd, Co, Cr, Cu, Hg, K, Mn, Mo, Ni, Sb, V, Zn</i>	<i>Cl⁻, SO₄²⁻, Na⁺, K⁺, Ca²⁺, Mg²⁺, EC</i>	<i>OC</i>

* Represent profile for PM₁₀ fraction; ** Bold font for present study; *** Italic font for reported international studies and; **** Italic font with underline for reported Indian study.

Table 5. Summary of chemical species abundance in PM_{2.5} emissions from industrial burning practices and their comparison with nationally and internationally reported profiles.

Source ID	Various studies	Chemical abundances in percent mass		
		< 0.1%	0.1–1%	1–10%
CPI	Present study	As, Cd, Co, Cr, Hg, Mn, Mo, Ni, Pb, Sb, Se, V, Cl, NO₃, NH₄⁺, K⁺, Mg²⁺	Cu, K, S, Zn, F, EC	Al, Fe, Mg, Na, SO₄²⁻, Na⁺, Ca²⁺, OC
	<i>Chow et al., 2004</i>	<i>As, Mg, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, V, Zn</i>	<i>Al</i>	<i>Ca, K, S, SO₄²⁻, K⁺, OC</i>
FEMNI	Present study	As, Cd, Co, Hg, Mo, Ni, Pb, S, Sb, Se, V	Cr, Cu, K, Zn, F⁻, Cl⁻, NO₃⁻, SO₄²⁻, K⁺, Ca²⁺, Mg²⁺	Al, Ca, Fe, Mn, Na, Na⁺, NH₄⁺, EC
SPI	Present study	As, Cd, Co, Hg, Mo, Ni, Pb, S, Sb, Se, V, NO₃⁻, NH₄⁺	Al, Cr, Cu, Mg, Mn, Na, Zn, F⁻, SO₄²⁻, Na⁺, K⁺, Ca²⁺, Mg²⁺	Ca, K, Cl⁻, OC
CTPP	Present study	Cd, Co, Cr, Hg, Mo, Ni, Pb, Sb, Se, V	As, Cu, Mg, Mn, Na, S, Zn, Cl⁻, NO₃⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺	Al, Ca, Fe, K, F⁻, SO₄²⁻, Ca²⁺, OC, EC
	<i>Watson et al., 2001</i>	<i>Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, V, Zn</i>	<i>As, Cl, K, Mg, Na, NO₃⁻, K⁺</i>	<i>Al, Ca, Fe, S, Cl⁻, NH₄⁺, OC, EC</i>
SRM	Present study	Al, As, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Sb, Se, V, F⁻, K⁺	As, Cr, Mg, Na, V, Zn, Cl⁻, NO₃⁻, Na⁺, K⁺, Ca²⁺	Ca, Ni, NH₄⁺, EC
	<i>CPCB, 2008</i>	<i>Al, As, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Sb, Se, V, F⁻, K⁺</i>	<i>K, Zn, Cl⁻, NO₃⁻, Na⁺, NH₄⁺, Mg²⁺</i>	<i>Ca, Fe, Mg, Na, Pb, SO₄²⁻, Ca²⁺, EC</i>
EAW	Present study	Cd, Co, Cr, Hg, Mo, Ni, Pb, Sb, Se, V, NO₃⁻	As, Cu, Pb, F⁻, Cl⁻, SO₄²⁻, Na⁺, NH₄⁺, Ca²⁺, Mg²⁺, EC	Al, Ca, K, Mg, Mn, Na, K⁺, OC
	<i>CPCB, 2008</i>	<i>As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Sb, Se, V, NO₃⁻</i>	<i>Na, Pb, F⁻, Cl⁻, SO₄²⁻, Na⁺, NH₄⁺, Ca²⁺, Mg²⁺, EC</i>	<i>Al, Ca, Mg, K, Mn, Zn, K⁺, OC</i>
BKP	Present study	Al, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, S, Sb, Se, V, Zn, F⁻	As, Fe, K, Mg, Na, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺	Ca, Ca²⁺
	<i>USEPA Speciate 4.3 PR. No.-91171</i>	<i>As, Cr, Cu, Mn, Ni, Pb, Sb, Se, Zn, NH₄⁺, EC</i>	<i>Cl, K, Na, Mg, S, NO₃⁻, SO₄²⁻</i>	<i>Al, Fe, OC</i>

* Represent profile for PM₁₀ fraction; ** Bold font for present study; *** Italic font with underline for reported international studies and; **** Italic font with underline for reported Indian study.

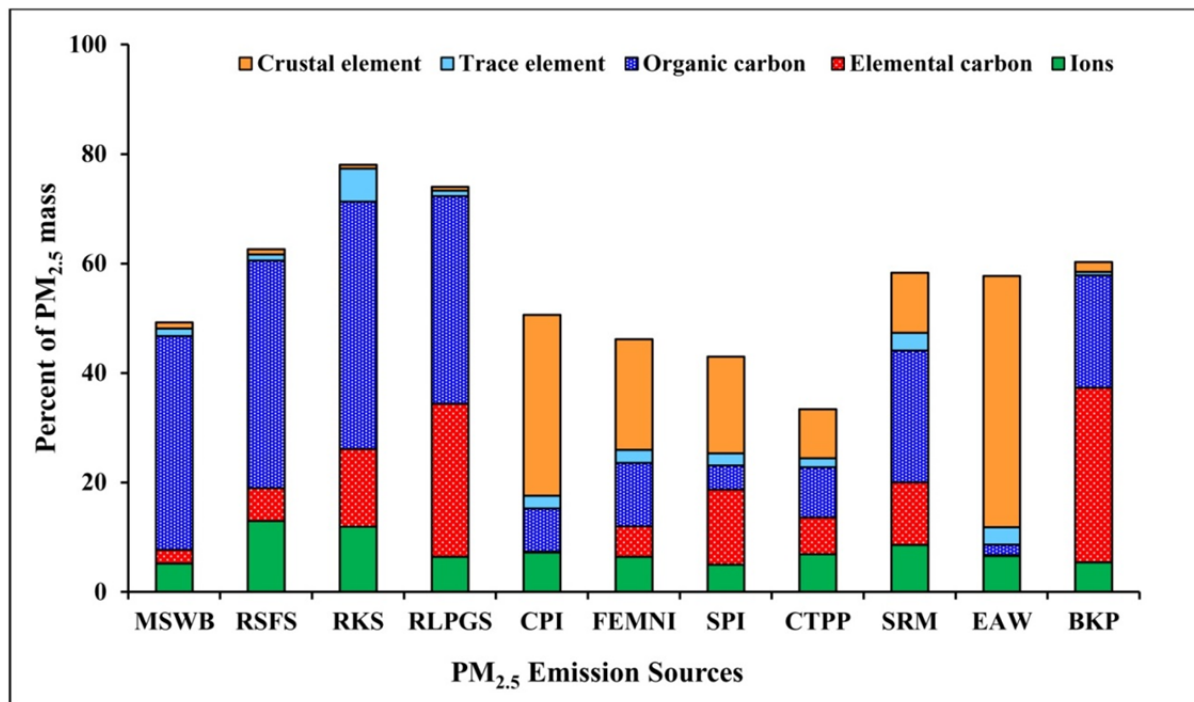


Fig. 2. Crustal elements (Al, Ca, Fe, K, Mg, Mn and Na), trace element (As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, S, Sb, Se, V, and Zn), ions (F^- , Cl^- , NO_3^- , SO_4^{2-} , Na^+ , K^+ , Ca^{2+} , Mg^{2+}) and organic content in $PM_{2.5}$ of selected source emissions.

Table 6. Source signatures of domestic and industrial burning practices in India.

Source type	Source signature	Source type	Source signature
	<i>Residential fuel burning</i>		<i>Industrial burning Sources</i>
RSFS	F^- , As, Mg^{2+} , Ca^{2+} , Cr, K^+	CPI	S, Cr, Cu, Al, Mo, Mg
RKS	Pb, Cd, Sb, F^- , Se, V	FEMNI	Cr, Mo, Mg^{2+} , Mn, NO_3^- , Cu
RLPGS	Sb, Cd, Pb, S, Mo, Se	SPI	Cr, As, Mg^{2+} , Cu, EC, Cl^-
<i>Residential fuel combustion^{a*}</i>	<u>OC, EC, K^+, Cl^-</u>	CTPP	As, Cr, S, F^- , NO_3^- , Al
	<i>Municipal waste burning</i>	SRM	Pb, Mg^{2+} , NO_3^- , EC, S, Cl
MSWB	F^- , Co, Cd, Ca^{2+} , Na^+ , K^+	EAW	Mn, Cr, K^+ , Cd, Pb, Mg^{2+}
<i>Waste burning^a</i>	<u>OC, EC, K^+, As, Pb, Zn</u>	BKP	As, Cd, Mo, EC, NO_3^- , Sb
		<i>Metallurgy^{a, b, d}</i>	<u>Mn, Zn, Pb, Cd, Cu, As, Hg</u>
		<i>Industry^{a, c} and Coal</i>	<u>V, Ni, SO_4^{2-} and Se, As, Cr, Co,</u>
		<i>Combustion^b</i>	<u>Cu, Al</u>

* Reported sources and their markers in italic style with underline for comparison.

^a Watson *et al.*, 2008; ^b Mitra *et al.*, 2002; ^c Viana *et al.*, 2008; ^d Viana *et al.*, 2006.

five-fold higher compared to RKS and RLPGS, respectively. On considering the biogenic markers (K^+ and NH_4^+), higher values of K^+/K ratio (0.89–0.97) and NH_4^+ abundance compared to those found in crustal origin and industrial emitted $PM_{2.5}$ is attributed to the fact that biogenic sources are dominating in $PM_{2.5}$ emissions from household solid fuel burning practices involved with cooking purposes. These values are similar to earlier reports on emissions of biogenic (Watson *et al.*, 1994, 2001; Chow *et al.*, 2004) and solid fossil fuel combustion (Watson *et al.*, 2001; Kong *et al.*, 2011). About 4–13 times higher K^+ emissions has been observed in case of RSFS compared to RKS and RLPGS, respectively. But NH_4^+ was found 2–16 times higher in emissions resulting from kerosene stoves (RKS) compared to RLPGS and RSFS, respectively. As far as trace species

is concern, sulphur constituents (S, SO_4^{2-}) along with Se and seven toxic species (As, Cd, Hg, Ni, Pb, V and Zn) were found multi-fold higher in RKS compared to RSFS and RLPGS; similar to earlier reported levels for kerosene stoves (Patil *et al.*, 2013). Other anions (NO_3^- , F^- and Cl^-) have shown higher presence in $PM_{2.5}$ emissions resulting from RSFS compared to RKS and RLPGS. Most of markers of crustal origin are found < 0.1% in all types of household fuel burning emissions.

Coal Fired Mineral Based Industries

Six different process based industrial emissions, mostly dominating in study region, have been chosen for the development of $PM_{2.5}$ chemical source profiles. Out of six chosen industries, only BKP has shown > 50% load of

carbonaceous matter (TC) in their PM_{2.5} emissions. Other industrial PM_{2.5} emissions have shown lesser load of TC in the ranges of 8.03–35.53%. PM_{2.5} emissions from two industrial combustion processes (SPI and BKP) have shown higher load of EC compared to OC with EC/OC ratio of 3.25 and 1.55, respectively. Significantly lower measured mass in case of CPI, FEMNI and CTPP is might be due to higher unaccounted silica load in PM_{2.5} mass. As far as iron particle load in PM_{2.5} is concern, sponge iron processing (direct reduction iron processing) (SPI) have shown 2–34 times higher load compared to other industrial processes included in the study. In case of other major constituents like Al is found highest in CTPP (15.5% of measured mass) and Ca is found highest in CPI (39.6% of measured mass). Water soluble/total ratio (Ca²⁺/Ca, Mg²⁺/Mg and Na⁺/Na) describes the pre-dominance of emissions resulting from either dust re-suspension during process handling or combustion activities involved with mineral based coal-fired industries (Volkovik, 1983; Watson *et al.*, 1994). The lower values of these ratios in PM_{2.5} emissions from CPI (0.13, 0.01 and 0.33, respectively), FEMNI (0.12, 0.25 and 0.58, respectively) and SRM (0.42, 0.31 and 0.18, respectively) attributed the higher abundance from crustal origin, whereas higher values for SPI (0.71, 0.55 and 0.61, respectively), CTPP (0.69, 0.73 and 0.68, respectively) and BKP (0.69, 0.28 and 0.81, respectively) indicated the pre-dominance of emissions from combustion processes. CTPP, SPI and BKP have shown higher enrichment of biogenic potassium (K⁺) with K⁺/K ratio of 0.73, 0.64 and 0.62, respectively; contrast to CPI and SRM along with earlier reported values for coal fired steel industries (Watson *et al.*, 2001; CPCB, 2008; USEPA, 2013). This might be due to use of poor quality coals in CTPP and SPI and biomass (dung cakes with coals) in brick kilns. Order of variation in association of anions with PM_{2.5} emissions from selected source sites is evaluated to be: NO₃⁻ > F⁻ > Cl⁻ > SO₄²⁻. On other hand, uniformity in occurrence of sulphur group species (S, SO₄²⁻ and Se) across the selected industrial sites is attributed to similar source origin of coal combustion (Volkovik, 1983). Trace elements have also shown significant variation in their relative abundances in PM_{2.5} emitted from different sources; justified their inclusion in the development of PM_{2.5} chemical source profiles for emissions resulting from combustion processes involved with different mineral based coal-fired industries. Brick Kiln processes (BKP) has been identified as the major emitter of highest number of toxic species (Cd, Co, Mo, Sb and V) followed by SRM (Hg and Pb) and SPI (As, Ni). Moderate combustion temperature and poor quality of coals and other combustion materials (dung cakes etc.) are responsible for higher emission of toxic species (Vollkovic, 1983).

In addition to these industrial sources, open fabrication workshops having arc-welding activities (EAW) is also reported to be the significant contributor of outdoor PM_{2.5} due to their profuse locations within the study region (Pervez *et al.*, 2005). It has been observed that EAW is contributing mainly active iron and manganese particles. Pb, As, Cu and F⁻ were observed to be found above 0.1% compared to other toxic species similar as earlier reported profiles of arc-

welding workshops (Swamy *et al.*, 1994; CPCB, 2008).

Comparison of Developed Source Profiles with Reported Profiles of National and International Origin

To compare the developed PM_{2.5} chemical source profiles in this study with previous reported profiles by Chow *et al.* (2004), Watson *et al.* (2001), the Central Pollution Control Board, India (2008) and Speciate 4.0 (USEPA). All chemical species of each of selected profiles are grouped in four percentage fractional ranges (Chow *et al.*, 2003, 2004) and summarized in Table 4 and Table 5. The major markers (OC, K⁺, NH₄⁺) of emissions resulting from MSWB were observed to be found in different levels in developed and reported profiles. OC was found > 10% in present and CPCB profiles, but that was observed to be within 1–10% in Speciate 4.0 profiles. K⁺ is found in different fractional range between present and CPCB profiles with higher abundance (1–10%) in presented profile; confirm the strong variation in biogenic matter content in MSWB. Additionally, the different abundance of K⁺ in MSWB emitted PM_{2.5} between present and CPCB profiles might be due to different sampling methods; CPCB adopted laboratory scale study, whereas real-world sampling on open air burning was adopted in present study. Apart from major markers, other species of crustal origin were found in similar fractional ranges in MSWB profiles developed in present study and CPCB database as well. In case of PM_{2.5} profiles of emissions resulting from residential fuel burning (RSFS, RKS and RLPGS), carbonaceous matter was found > 10% across all the comparative profiles, but EC was found > 10% only in presented profiles. As far as anions and cations of fuel combustion markers (K⁺, NH₄⁺, SO₄²⁻, Cl⁻, NO₃⁻) is concern, most of them found higher (1–10%) in presented profiles compared to databases of CPCB and Speciate 4.0; most of them found higher in emissions of kerosene stoves (RKS). Other species of crustal origin and trace elements have shown more than 50% agreement in their occurrence in similar fractional groups across the selected databases for comparison.

On comparing PM_{2.5} chemical profiles of emissions resulting from selected industrial sites with CPCB and Speciate 4.0, different observations were obtained for different industrial sites. Sponge-iron industrial emissions (SPI) and Ferro-manganese industry (FEMNI) were not included in CPCB and Speciate 4.0. In case of CPI and CTPP, major fractional group (> 10%) has shown different inclusion of species across the comparative databases. BKP has shown inverse distribution of OC and Ca between two fractional groups (> 10% and 1–10%) on comparing the presented profile with CPCB profile. On contrary, EAW has shown similar major marker (Fe) in both comparative source profile databases. The element Ca for CPI, OC and EC for SRM & BKP, Fe for EAW, EC for SPI and OC for FEMNI were accounted for > 10% abundances. The water soluble ions and crustal element distributed between 0.1–1% and 1–10% ranges. The trace elements were contributed < 0.1% relative abundances except As and S for CTPP with range 0.1–1%.

Overall species abundance of source profiles in defined

fractional groups is in ~60–80% agreement with reported source profile databases of CPCB and in ~35–50% agreement with USEPA Speciate 4.0.

Mass Closure Analysis

Mass reconstruction of PM_{2.5} has been carried out using revised IMPROVE18 mass closure (material balance) equation (Ho *et al.*, 2003, Chow *et al.*, 2012); estimate the unmeasured oxides and compared with the total gravimetric measured mass for the quality assurance (Watson *et al.*, 2012):

$$[PM_{2.5}] = 1.375 SO_4^{2-} + 1.29 NO_3^- + 1.8 OC + EC + (2.2 Al + 2.49 Si + 1.63 Ca + 1.94 Ti + 2.42 Fe) + 1.8 Cl^- \quad (1)$$

Two major crustal elements, silica and titanium have not been included in mass closure study; resulting in lower estimated values for crustal fraction in the overall mass closure. In case of RKS and RLPGS, PM_{2.5} mass reconstruction has been achieved 107.27% and 102.08%, respectively due to negligible abundances of unmeasured silica, titanium and higher abundance value of OC. Domestic solid material burning practices (RSFS and MSWB) have shown relatively lower mass closure values (97.47% and 79.36%, respectively). PM_{2.5} Mass closure results for industrial sources have shown close equivalence between the gravimetric mass and reconstructed mass. The highest and lowest mass closure values have been found in case of EAW (98.84%) and CTPP (45.65%), respectively. Mass closure results of PM_{2.5} emissions, resulting from other industrial source sites, were achieved 77.13%, 62.11%, 66.32%, 68.12% and 85.08% for BKP, FEMNI, SPI, CPI, and SRM, respectively. No overestimation of mass closure result has been occurred in developed profiles.

Ion Balance Calculation

The ionic balance calculation has been performed to confirm the acid-base property of PM_{2.5} fractions emitted by different burning practices. Conversion of ion mass concentrations into micro equivalents was performed to calculate the cation/anion balance of PM_{2.5} (Cao *et al.*, 2005; Zhang *et al.*, 2011; Tao *et al.*, 2013). The cation and anion micro equivalents of particles were calculated as follows:

$$C \text{ (Cation micro equivalents/m}^3\text{)} = Na^+/23 + NH_4^+/18 + K^+/39 + Mg^{2+}/12 + Ca^{2+}/20 \quad (2)$$

$$A \text{ (Anion micro equivalents/m}^3\text{)} = F^-/19 + Cl^-/35.5 + NO_3^-/62 + SO_4^{2-}/48 \quad (3)$$

The well balanced anion/cation (A/C) ratio must be 1. The value higher than 1, indicates the acidic nature of the particle (Kerminen *et al.*, 2001) whereas slightly lower than 1, indicate contribution of unmeasured CO₃²⁻ ion, and very low A/C ratio indicates the basic nature of particle (Cao *et al.*, 2005; Shen *et al.*, 2007, 2009). A/C ratios of PM_{2.5} emissions resulting from domestic and industrial burning practices have been accounted; ranges from 0.18–0.79 and 0.19–0.74, respectively and confirm that all source emitted

PM_{2.5} samples were basic in nature.

Source Markers

Source markers 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). The relative source indicator species were evaluated for all eleven sources grouped in domestic and industrial burning practices. For the calculation, following formula was applied to define the indicatory species for specific source emitted PM_{2.5} fraction (Yang *et al.*, 2002; Kong *et al.*, 2011):

$$Ratio_{i,j} = \frac{(X_i / \sum X)_j}{(X_i / \sum X)_{\min}} \quad (4)$$

where: X_i was the ith individual species concentration; (X_i/∑X)_j was the quotient of ith individual species ij divided by the summation of 32 species concentrations of emission source j; (X_i/∑X)_{min} was the quotient of ith individual species divided by the summation of 32 species concentrations which were the minimum for all emission sources (Yang *et al.*, 2002; Chen *et al.*, 2003). A normalization procedure has been applied according to Mitra *et al.* (2002) and Kong *et al.* (2011) to minimize the effect of physical parameters. Normalized individual species concentration was used by dividing the ith individual species ij concentration to the sum of ith individual concentration for all the source profiles (Kong *et al.*, 2011). The top six chemical species with highest ratio values for total relative source profiles has been used as relative source indicatory chemical species describe in detail on Table 6 and compare with earlier reports (Kong *et al.*, 2011). Water soluble K⁺, marker of vegetative and biomass burning sources (Watson *et al.*, 2002, 2008), F⁻ and Ca²⁺ were evaluated to be the similar source markers of PM_{2.5} emissions from burning practices involved with MSWB and RSFS; whereas RKS and RLPGS have shown distinct source markers with major marker groups of (F⁻ and V) and (S and Mo), respectively. Arsenic (As) was found common source marker of PM_{2.5} emissions from CTPP, SPI, and BKP; one of prominent trace element marker of emissions resulting from coal burning (Mitra *et al.*, 2002). Mn was calculated as common source marker PM_{2.5} emissions from FEMNI and EAW. Distinct observations of source markers of PM_{2.5} emissions from selected source sites compared to those reported earlier (Mitra *et al.*, 2002; Viana *et al.*, 2006; Watson *et al.*, 2008) is attributed to the importance of development of region specific source profiles to obtained precise results of receptor modeling.

CONCLUSIONS

The differences with earlier reported/developed similar characteristics profiles created demand of additional and more precise source profiles that represent a study area. In this channel, the present study is an important work in development of source profile database in India. The eleven important stationary sources profiles for PM_{2.5} fraction are

reported in this paper. These profiles are comprised of 21 element, 9 water soluble ion, and carbonaceous fractions by following the standard protocol of chemical analysis and data validation. The carbonaceous fractions are most abundant with different OC/TC ratio; ranges from ~0.58–0.94 in PM_{2.5} from selected source emissions. Trace metals were found significantly higher in PM_{2.5} emissions from burning practices involved with household cooking activities and municipal solid waste management practices, compared to crustal origin. Observation of different relative enrichment of defined chemical components (Fig. 2) in PM_{2.5} emissions from selected industrial burning sources might be due to use of different raw materials, and combustion temperature and conditions involved with industrial processes. The developed profiles comparatively much similar in > 10% and < 0.1% abundant species with earlier reported profiles for similar sources. The K⁺, Mn, and As were found and source marker for biomass burning, metallurgical industrial emission, and coal burning respectively, shown good agreement with National CPCB, 2008 and global USEPA speciate database also previous reported profiles.

These profiles require update, up gradation and addition of new sources with the sufficient interval of time to better represent changes in characteristics of sources of burning practices in India.

AUTHOR CONTRIBUTIONS

S.P. designed and led the study, including project coordination; S.P. and J.M. were involved with field measurements, data collection, manuscript preparation and data analysis; J.M., S.D., S.T. and D.B. performed Chemical analysis. All authors discussed the results and contributed to the manuscript.

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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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Supplementary Materials

Table S1. Instrument parameters with filter blank and field blank values.

Instrument	Species	Wavelength	Calibration	LOD in ppb	Blank (g/Filter)	Field blank (g/Filter)
AAS-Flame	Ca	422.7	0.999	0.0037*	0.000010	0.000010
	Fe	248.3	0.999	0.0043*	0	0.000007
	K	766.5	0.999	0.0009*	0.000002	0.000007
	Mg	285.2	0.999	0.0022*	0.000003	0.000002
	Na	589.0	0.994	0.0037*	0.000024	0.000034
	Zn	213.9	0.999	0.0033*	0.000007	0.000010
AAS-GF/Zeeman	Al	309.3	0.978	0.21	0	0.000099
	Cd	228.8	0.999	0.02	0	0
	Co	240.7	0.998	0.01	0.00000004	0.00000005
	Cr	357.9	0.998	0.025	0.00000009	0.0000003
	Cu	324.8	0.993	0.29	0.0000004	0.0000007
	Mn	279.5	0.962	0.06	0.00000002	0.0000010
	Mo	313.3	0.981	0.31	0.0000002	0.0000004
	Ni	232.0	0.999	0.16	0.0000002	0.0000021
	Pb	283.3	0.982	0.07	0	0
	V	318.5	0.998	2.7	0.00000003	0.0000002
	As	193.7	0.999	0.53	0	0.000000009
	Sb	217.6	0.999	0.4	0.0000000008	0.000000005
	Se	196.0	0.999	0.8	0.00000002	0.00000002
AAS- VP Spectrophotometer	Hg	253.7	0.999	0.06	0.0000002	0.00000059
	NH ₄ ⁺	620	0.998	-	0.0000025	0.00000774
Ion chromatograph	F ⁻	-	-	0.01*	0	0.0000185
	Cl ⁻	-	-	0.01*	0.0000643	0.000103
	NO ₃ ⁻	-	-	0.01*	0	0
	SO ₄ ²⁻	-	-	0.01*	0.000027	0.000044
SemiContinuous Carbon analyzer	OC	-	-	-	0.000109	0.000176
	EC	-	-	-	0.0000000037	0.000000203

*LOD value in ppm unit.