



Impact of Different Household Fuel Use on Source Apportionment Results of House-Indoor RPM in Central India

Shamsh Pervez^{1,2*}, Neha Dubey¹, John G. Watson², Judith Chow², Yasmeen Pervez³

¹ School of Studies in Chemistry, Pt. Ravishankar Shukla University, Raipur, CG, 492010, India

² Division of Atmospheric Sciences, Desert Research Institute, 2215 Raggio Parkway, Reno, NV, 89512, USA

³ Dept. of Engineering Chemistry, CSIT, Kolihapuri, Durg, C.G., 491001, India

ABSTRACT

Conventional character of household fuel use in India showed significant impact on residential-indoor air quality and consequently deteriorating personal air quality of inhabitants. Multi-complexity in location and type of industrial units along with haphazard traffic system are some of the major challenges in resolving relative source contribution with higher significance, especially in case of indoor and personal level air quality. On the basis of questionnaire survey that include daily activity pattern with use of household cooking fuel by local residents, three fuel categories identified: 1) Use of Liquid Petroleum Gas (LPG) stove, 2) Use of kerosene stoves and electric stoves and 3) Conventional stoves using coke and cow dung cakes. In many of middle class residents, kitchen cooking is depending on all three categorized fuels. A longitudinal measurements of respirable particulate matter (RPM) in residential-indoors, ambient-outdoors of selected source sites and local-outdoor of road-traffic junctions have been monitored during October–December 2009. Sampling frequency was ten (twice in a week). Chemical Mass Balance (CMB8) was executed using source-routes and indoor-receptor compositional profiles. Results have shown significant variation in relative contributions of outdoor sources with potential impact of household fuel emission on source contributions of indoor RPM. Results also explained extent of contribution from road-traffic emissions, local soils and major industrial complex to indoor RPM levels.

RELEVANCE: The study is the part of comprehensive assessment of source apportionment of particulate matter at ambient, indoor, personal and surface levels in urban-industrial environment of central India using 21 chemical species have been analyzed using standard reported protocol of ICP-AES and ion-chromatograph. Source apportionment has been conducted using two methods: Preliminary- linear regression, then using CMB8.

Keywords: Source apportionment; Indoor air; Respirable particulate matter; Household fuel.

INTRODUCTION

Unprocessed solid fuels such as coal, wood, and animal waste are combusted for cooking and heating by low-income populations in developing countries (Raiyani *et al.*, 1993; Ge *et al.*, 2004; Pohekar *et al.*, 2005; Farsi *et al.*, 2007; Basu *et al.*, 2008; Joon *et al.*, 2009; DeFries and Pandey, 2010). This Household Fuel Burning (HFB) elevates indoor and outdoor pollutant concentrations and human exposures, with a large variation depending on the fuels, burning methods, and appliances. Excessive exposure to these emissions is associated with several adverse health effects, especially for women and children (Bhargava *et al.*, 2004; Pokhrel *et al.*, 2005; Kanishtha *et al.*, 2006; Padhi and Padhy, 2008; Mondal *et al.*, 2010).

However, solid fuel combustion is only one contributor to excessive Respirable Particulate Matter (RPM, particles with aerodynamic diameters $< 5 \mu\text{m}$) in indoor environments (Gadkari and Pervez, 2007, 2008). Infiltration of outdoor air, re-suspension of house dust, bio-aerosols from plants and animals, and many other sources can also be large contributors to indoor air (Sexton and Hayward, 1987; Adgate *et al.*, 1998; Hidy *et al.*, 2000; Kopperud *et al.*, 2004; Larson *et al.*, 2004; Cao *et al.*, 2005a; Cao *et al.*, 2005b; Hu *et al.*, 2006; Gadkari and Pervez, 2007; Cortez-Lugo *et al.*, 2008; Gadkari and Pervez, 2008; Hu *et al.*, 2009). The purpose of this study is to quantify contributions from different sources to indoor RPM levels in an urban-industrial region of southeast-central India, with special attention to the HFB source. This is accomplished by measuring elemental markers for different source types and interpreting them using receptor models.

* Corresponding author.

E-mail address: shamshp@yahoo.co.in;
shamshpervez@gmail.com

METHODOLOGY

Indoor-receptor Sampling

Measurements were obtained from October 02 through December 05, 2009 at four homes in Raipur, Chhattisgarh, India, as shown in Fig. 1. Sampling sites were selected according to established criteria (Gilbert, 1987; Abt *et al.*, 2000; Crist *et al.*, 2008; Begum *et al.*, 2009; Massey *et al.*, 2009) that describe the following parameters have to be included in decision of sampling sites: meteorological records (wind direction, wind velocity, relative humidity and average rainfall), layout map, development plan, population density and human activity pattern, soil texture and hydrology, representation from environmentally defined urban receptors (pure residential, commercial and sensitive zones), and epidemiological pattern of the study area. Windrose of the study area has been presented in Fig. 2. Indoor monitoring sites have been identified within the established ambient

air monitoring network before (Dubey and Pervez, 2008). Table 1 describes the characteristics of the indoor and outdoor environments. Handy respirable particulate samplers (Envirotech Model APM 821) drew air at 1.1 L/min over 24-hour periods through cyclones with a 5 μm 50% cut-point onto 37 mm diameter glass-fiber filters (Whatman GF/F). One sampler was placed inside each residence and two samplers were located outside, one 500 m from the residence to represent outside air (especially from road-traffic junction) that might infiltrate and the other 2500 to 3500 m from the residence next to the closest emissions sources, as detailed in Table 1. Two samples per week were taken over a ten-week period in each of the four areas shown in Fig. 1. In each sampling event three GFF's were recovered, each in an interval of 8 hrs to overcome the filter choke and to maintain the uniform air flow rate.

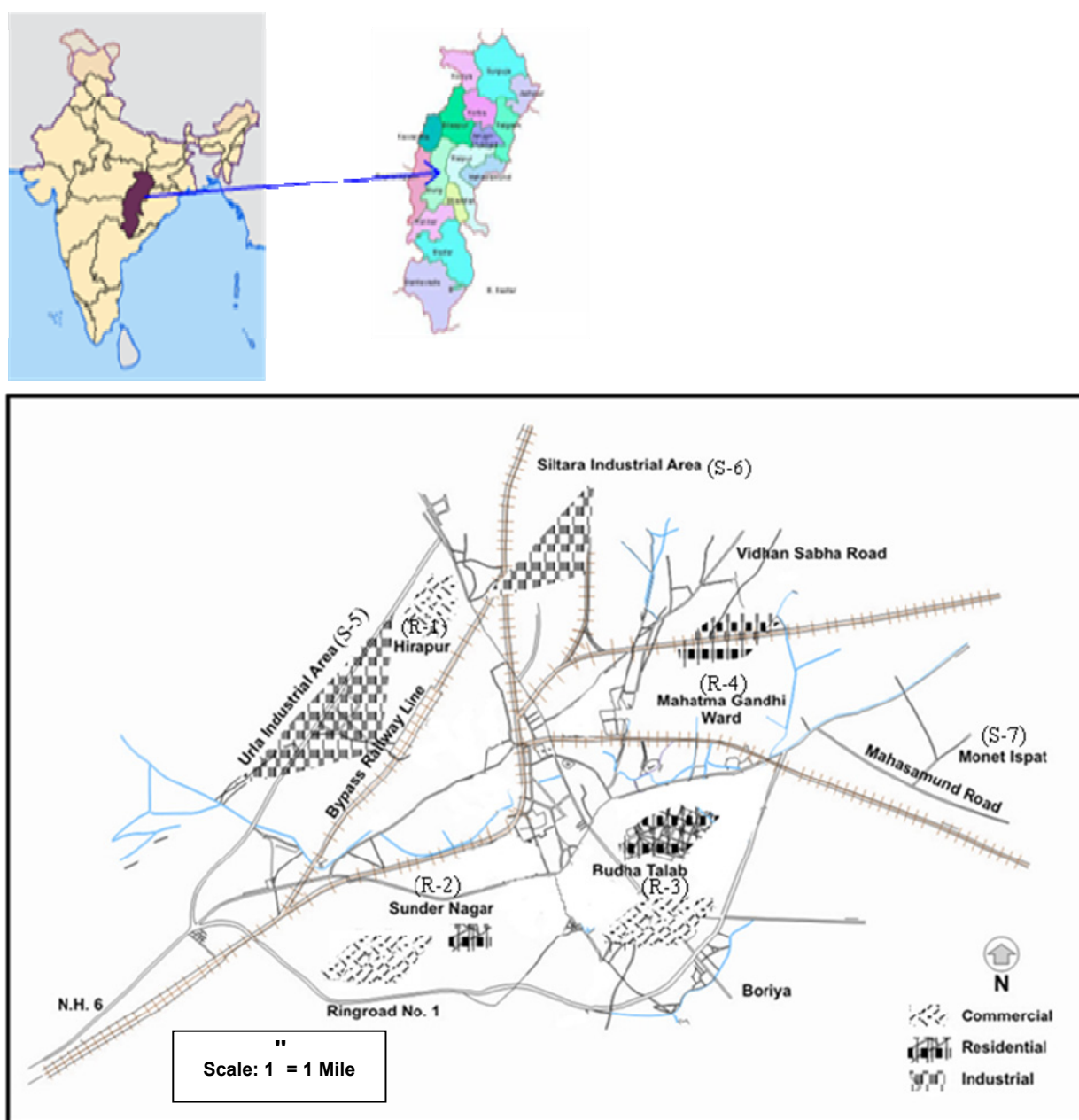


Fig. 1. Locations of residences and outdoor-sources in Raipur, CG, India.

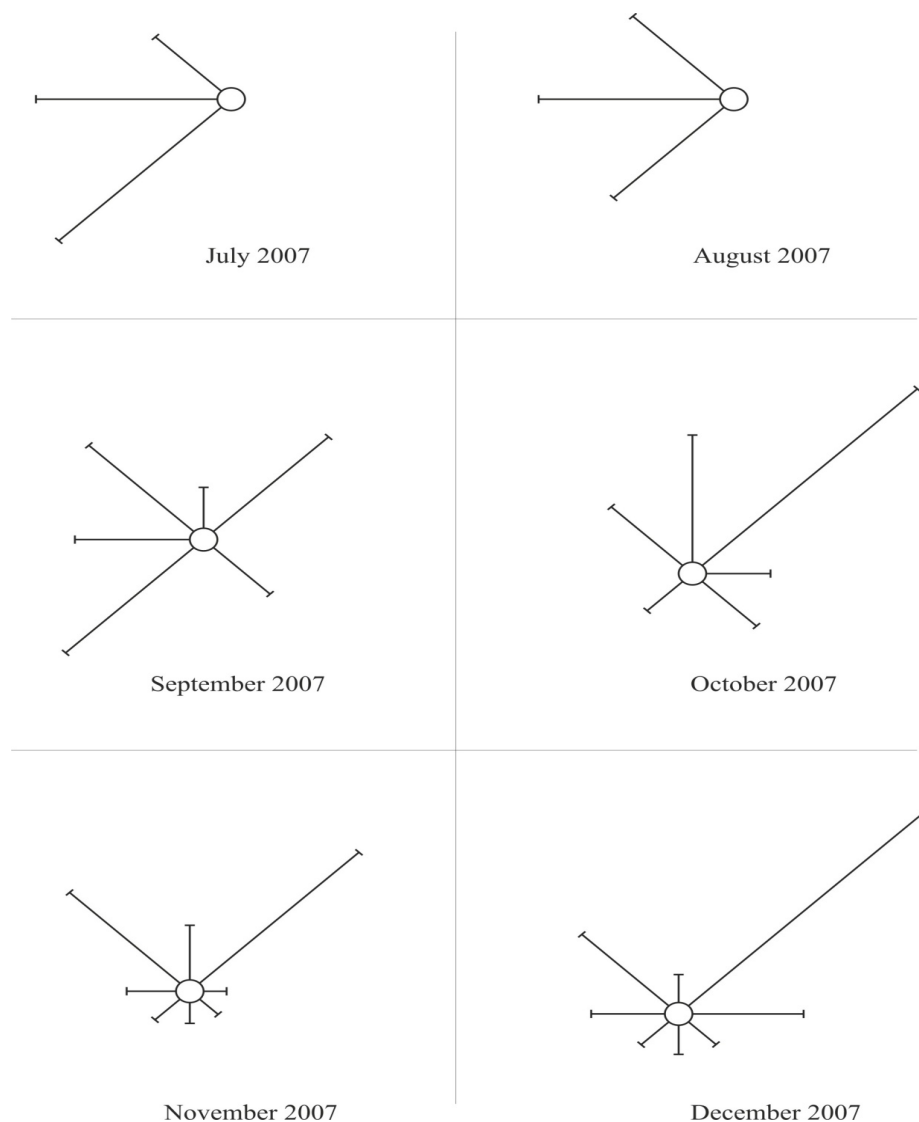


Fig. 2. Wind rose of the study region during July- December 2007 (sampling period was October–December 2009).

Source Sampling

Source emissions samples were acquired for different residential fuels to develop elemental source profiles for these emissions, as summarized in Table 2. Fuels were combusted in three different stoves for 8 to 12 hours within a closed room in and sampled with the same sampler used for outdoor and indoor air. Similar conventional stoves have been widely used across the local community.

For fugitive dust profiles, 10 kg of surface soils were obtained from surfaces near the monitoring sites. For the study region as a whole, the predominant soil type is clayey soil. Distribution of different soil type within study region has been found as: Clayey- 45.94%, Laterite- 14.26%, Sandy loamy- 21.23% and clay-loamy- 18.57%. Soil samples representing all available types have been collected from five locations close to indoor sites using standard procedures (Koshle *et al.*, 2008) in a depth of 4–6 cm after removing upper surface layer. Collected soils were then mixed together in similar proportion described as relative availability in the study region. These samples were ground in a milling

machine and suspended into a glass chamber (size 1.5 m³) using a pressure fan from which they were sampled onto filters using the same Handy sampler and inlet (Chow *et al.*, 1994; Gadkari and Pervez, 2007). In each sampling event three GFF's were recovered, each in an interval of 4 hrs to overcome the filter choke and to maintain the uniform air flow rate.

Filter Analysis

Before and after sampling the filters were conditioned in desiccators at 55°C and 55% RH for 12 hours prior to weighing on an electronic balance (Denver Instrument Model TB-215D) with 0.01 mg precision. The net weight gain was divided by the sample volume to obtain RPM concentrations in µg/m³ at ambient temperatures and pressures.

A 3.5 cm² portion of each of three exposed filter in each sampling (total:10.5 cm²) plus filter blanks was extracted in 50 ml of deionized distilled water and submitted to ion chromatography for Cl⁻, SO₄²⁻, and NO₃⁻ levels (Canepari *et al.*, 2006). Another 10.5 cm² portion was extracted in 25

Table 1. Description of Sampling Sites for indoor/outdoor RPM monitoring within an urban-industrial area of Raipur, CG, India.

S.No.	Name of site and Code	Site description							Sampling site
		Ambient/Indoor-Receptor site							
		Outdoor surroundings			Indoor environment				
		Location type	Outdoor roads	Locality	House type	Ventilation	Fuel use		
1	Hirapur Ward, Raipur (R-1)	Residential and small scale industries	Paved roads	Colony, no green land	2 BHK, Brick work	Moderate ventilation	LPG & kerosene	5 ft in living room	
2	Sunder-Nagar, Raipur (R-2)	Pure residential cum sensitive area	Paved roads	Colony, no green land	2 BHK, Brick work	Poor ventilated	Mix-LPG, kerosene and dung cake	5 ft in living room	
3	Budha-Talab, Raipur (R-3)	Commercial cum heavy traffic area	One side paved and two side unpaved road with high traffic	Colony, no green land	3 BHK, Brick work	Well ventilated	LPG & kerosene	5 ft in living room	
4	M.G. Ward, Raipur (R-4)	Close proximate to industrial complexes	Paved roads	Colony, no green land	1 BHK,	Moderate ventilation	LPG & kerosene	5 ft in living room	
Outdoor-Source site									
5	Urla industrial complexes (S-5)	Most of casting industries, chemical industries, medium scale workshops							
6	Siltara industrial complexes (S-6)	Most of sponge iron industries,							
7	Mandir Hasaud industrial complexes (S-7)	Medium steel industries.							
8	Road Traffic (S-8)	automobile exhaust & road-side runoff							
Additional sources for development of chemical profiles in CMB execution									
9	Soils (S-9)	Local origin (representative from all selected receptors)							
10	Residential Fuel (S-10)	Coal burning, LPG, Kerosene burning							

Table 2. Physical characteristics of stoves used for burning of selected household fuels for development of source profile.

Stoves characteristics, operated together				
Stove type	Dimensions	Fuel used	Height of stove burners	sampler's inlet height
Conventional stove	cylindrical (ht: 2.5 ft, dia: 1.5 ft)	3.5 kg of coke and cow dung cakes (3:1) by wt	2.5 ft	3.5 ft
LPG stove	Rectangular (3 ft × 1.5 ft × 0.5 ft)	LPG	2.5 ft (Adjusted)	3.5 ft
Kerosene stove	cylindrical (ht: 1 ft, dia: 0.65 ft)	Kerosene	2.5 ft (Adjusted)	3.5 ft

mL acid mixture of H₂O₂ and HNO₃ (3:1) by standard digestion method applied for 6–8 hrs (Canepari *et al.*, 2006; Tursic *et al.*, 2008). Final volume was made up to 25 mL using deionized distilled water and submitted to ICP-AES analysis for Fe, Al, Ca, Mg, Cr, Mn, Ni, As, Hg, V, Zn, Cu, Pb, Co and Sb using standard procedures and recommended conditions of operation (Montaser and Golightly, 1987). A portion of this extract was analyzed by AA for Na and K. KCl and NaCl (Merck, Spectroscopic grade) were used for calibration of the instrument (Balakrishna and Pervez, 2009). Blank filter values were found < BDL (Below Detection Limit) in a calibration range from 0.001–1.0 ppm

of respective species concentration except for (µg/m³): TC (0.113); Cl⁻ (0.158); NO₃⁻ (0.0675); SO₄²⁻ (0.0234); Na (0.0993); K (0.0040); Ca (0.0180) and Mg (0.0123). A final 10.5 cm² portion was analyzed for total carbon by combustion and detection of the evolved CO₂ (Balakrishna and Pervez, 2009). Filter blanks were subtracted from the amounts on indoor and source samples prior to dividing by the sample volumes. Five replicate analytical measurements have been done to maintain relative standard deviation within 5%. Average and standard deviations of RPM levels and their chemical characteristics have been reported in Tables 3 and 4.

Table 3. Mean Indoor RPM levels ($\mu\text{g}/\text{m}^3$) and their correlation statistics with selected source oriented ambient RPM in an urban area.

Source site Receptor Site	S-5 (229.70 ± 107.60)*	S-6 (148.44 ± 29.89)	S-7 (118.07 ± 33.08)	S-8 (163.23 ± 46.24)
Correlation statistics (r)				
R-1 (417.89 ± 75.66)	−0.38	0.47	−0.11	0.16
R-2 (466.99±101.89)	−0.33	0.71	−0.44	−0.48
R-3 (351.91 ± 93.09)	−0.29	0.45	0.32	0.66
R-4 (754.27 ± 77.31)	−0.37	0.76	0.09	0.33

*Values in parenthesis: Annual average RPM level (Geometric mean ± standard deviation).

Table 4. Chemical characterization (Geometric Mean ± Standard Deviation) of Longitudinal Measurements of Indoor RPM.

Receptor	Concentration ($\mu\text{g}/\text{m}^3$)					
	TC	SO ₄ ²⁻	Cl ⁻	NO ₃ ⁻	Fe	Al
R-1	(125.148 ± 58.265)	(12.212 ± 11.347)	(17.090 ± 10.945)	(12.837 ± 5.776)	(17.796 ± 11.474)	(0.100 ± 0.043)
R-2	(110.811 ± 31.294)	(42.03 ± 9.941)	(0.013 ± 0.003)	(1.008 ± 0.739)	(55.957 ± 7.917)	(0.037 ± 0.014)
R-3	(103.04 ± 15.228)	(33.552 ± 3.494)	(9.457 ± 1.799)	(2.675 ± 0.597)	(40.854 ± 10.93)	(0.730 ± 0.484)
R-4	(158.039 ± 33.81)	(22.451 ± 14.996)	(22.504 ± 15.26)	(37.540 ± 13.448)	(52.810 ± 8.84)	(0.457 ± 0.169)

Receptor	Concentration ($\mu\text{g}/\text{m}^3$)					
	Na	K	S	Cr	Mn	Ni
R-1	(0.025 ± 0.009)	(0.378 ± 0.293)	(0.218 ± 0.092)	(0.257 ± 0.087)	(0.032 ± 0.040)	(1.724 ± 0.776)
R-2	(3.890 ± 0.883)	(23.387 ± 6.021)	(4.997 ± 0.90)	(0.0032 ± 0.003)	(0.000004 ± 0.000003)	(0.00018 ± 0.00015)
R-3	(4.603 ± 1.759)	(9.097 ± 2.208)	(3.957 ± 0.995)	(0.0006 ± 0.0005)	(0.018 ± 0.007)	(0.002 ± 0.0009)
R-4	(38.015 ± 15.136)	(15.065 ± 14.068)	(6.821 ± 2.73)	(0.030 ± 0.021)	(0.424 ± 0.404)	(20.106 ± 6.589)

Receptor	Concentration ($\mu\text{g}/\text{m}^3$)					
	V	Zn	Cu	Pb	Co	Sb
R-1	(0.062 ± 0.053)	(0.072 ± 0.072)	(0.017 ± 0.007)	(0.033 ± 0.013)	(0.0003 ± 0.001)	(0.0005 ± 0.0012)
R-2	(0.00004 ± 0.00003)	(0.004 ± 0.002)	(0.0001 ± 0.00005)	(0.002 ± 0.001)	(0.0001 ± 0.00014)	(0.0001 ± 0.0002)
R-3	(0.00006 ± 0.00002)	(0.003 ± 0.0034)	(0.009 ± 0.064)	(0.024 ± 0.008)	(0.004 ± 0.00001)	(0.045 ± 0.011)
R-4	(0.325 ± 0.261)	(62.397 ± 15.383)	(0.389 ± 0.335)	(60.231 ± 20.617)	(34.761 ± 13.856)	(7.853 ± 5.150)

Receptor	Concentration ($\mu\text{g}/\text{m}^3$)				
	Ca	Mg	As	Hg	UNDV
R-1	(0.261 ± 0.134)	(0.020 ± 0.009)	(0.291 ± 0.160)	(0.335 ± 0.158)	(156.332 ± 105.424)
R-2	(50.985 ± 17.292)	(1.875 ± 0.591)	(0.003 ± 0.0007)	(0.001 ± 0.0007)	(59.685 ± 116.991)
R-3	(24.694 ± 4.283)	(1.945 ± 1.169)	(0.001 ± 0.0001)	(0.0001 ± 0.0003)	(86.106 ± 93.34)
R-4	(0.223 ± 0.097)	(3.0008 ± 0.762)	(37.221 ± 26.731)	(11.559 ± 12.310)	(77.161 ± 96.376)

(UNDV = Undetectable portion of PM)

RESULTS AND DISCUSSION

Indoor/Outdoor Concentrations (Tables 3, 4 & 5)

Mean RPM levels of selected indoor-receptors have shown significantly higher presence compared to Indian NAAQS standards ($60 \mu\text{g}/\text{m}^3$ for residential areas). The annual average ratio RPM/PM₁₀ was found to be 0.69 (Gadkari and Pervez, 2007) and was being used for comparison of indoor RPM measurements with Indian NAAQS of PM₁₀. Results of regression analysis between indoor-receptors and outdoor-routes have shown higher degree of variation in relationship. In contrast to Urla Industrial area (S-5), Siltara Industrial area (S-6) has shown best correlation with all indoor-receptors compared to other source-routes. This might be due to favorable

wind direction for S-6 towards the indoor-receptor sites. The prevailing wind direction in the study region is mainly northeasterly and southwesterly throughout the year, but it was mostly southwest during the study period (October–December 2009). The deviation pattern from annual geometric mean can also be a marker to explain the negative correlation of S-5 with all indoor-receptors, which is unusually very high compared to other source-routes. The higher deviation pattern at R-5 is due to location of variety of industrial units having different manufacturing processes involved. The Mandir Hasaud Industrial area(S-7) has shown varied and poor relationship with all indoor-receptors. As far as Road-traffic (S-8) is concern, R-3 and R-4 have shown maximum susceptibility due to their surroundings of high traffic density and commercial zone. Highest correlation coefficient (0.75)

between S-6 and R-4 is due to their close proximity.

It has been clearly observed from regression analysis method (Geller *et al.*, 2002) that S-6 has shown major contribution to most of indoor-receptors starting from 53% to R-4, 36% to R-1 and 33% to R-3 and this is due to three factors: 1) favorable wind direction from this source site, 2) About 95% of industrial units are based on high temperature based metallurgical processes, which release RPM in a uniform pattern and 3) huge presence of iron and carbon particles that have been reported to be the good adsorbents of other chemical species and have high settling tendency within urban scale (Borderieux *et al.*, 2004). S-7 and S-8 have shown marginal contribution to selected sites with exception of R-3 by S-8. Occurrence of higher degree of variation in source-receptor relationship has implied the need to execute chemical mass balance with taking soils and HFB also as additional source-routes of indoor RPM.

The chemical analysis of indoor RPM has shown that source based marker species were found comparatively in greater levels than other species. Carbon and Iron content have been quantified at higher and uniform level across the selected residential indoors located in different surroundings and 4–7 folds higher than earlier indoor studies conducted in Indian environment (Thakur *et al.*, 2004; Rai *et al.*, 2002). This might be due to greater number of ferrous metallurgical industries located close proximity to study area. Anionic species (SO_4^{2-} , NO_3^- and Cl^-) have shown 2.5–10 times higher presence across all selected indoors compared to earlier reported values (Verma *et al.*, 2010; Kulshrestha *et al.*, 2009). All anionic species have shown different levels across the indoor receptors with higher levels in R-4 located close to Siltara industrial area. Higher presence of SO_4^{2-} in R-2, pure residential area located in clean environment, might be due to favorable wind direction from selected industrial locations during sampling period. As far as earth crustal species (Al, Ca and Mg) are concern, again different enrichment in RPM has been observed with higher Ca values at R-2 followed by R-3, while RPM-Al has shown higher presence at R-4 due to close proximity of S-6. Mg and Na have shown uniform increasing enrichment in RPM from R-1 to R-4. A potassium levels in RPM have been found to be higher in R-2 and R-4 compared to other indoors due to dominance of indoor generated RPM compared to outdoors. Interestingly, sulfur has shown lower trend in indoor RPM enrichment at R-1 due to closeness to S-5 where most of industrial units are non sulfur based chemical/fertilizer manufacturing and textile units. Chromium levels have shown presence within prescribed standards but R-1 and R-4 have received significantly higher concentration compared to earlier reported values. Manganese has shown comparable levels compared to earlier reported values. Nickel has shown different enrichment pattern in indoor RPM with hundred fold higher levels at R-1 and R-4 compared to prescribed standards, while R-2 and R-3 have not shown high RPM-Ni levels. As, Hg, V, Zn have shown uniform variation in RPM enrichment across residential-indoors with higher enrichment at R-1 and R-4 due to their close proximity to industrial locations, and hundred to thousand fold higher levels compared to prescribed standards.

Lead has shown hundred fold higher presence in RPM measured at R-4 indoors, while other sites have not shown higher RPM-Pb compared to prescribed standards. Similar trends of elemental enrichment in indoor-RPM have been observed in case of cobalt for R-4 and antimony for R-3 and R-4 both.

Source Profiles (Fig 3)

Statistical graph of \log_{10} of species concentration (%) have been presented in Fig 3. The logarithmic values of source species concentrations (%) have been taken to resolve the relative positions of statistical graphs of selected species. Results of source chemical profiles have shown that total carbon has been emerged as major constituent of RPM matrix in all sites except S-5, where major emissions from non-coal based non-metallurgical processes. Total carbon (TC) has shown major presence in road traffic emissions (S-8) followed by S-7, S-6 and S-10. S-6 and S-7 have 95% industrial units with higher temperature based combustion processes whereas most of industrial units located in S-5 region are known for moderate temperature based processes. Similarly higher deviation pattern in longitudinal measurements of TC at S-5, S-9 and S-10 compared to S-6 and S-7 is also due to process temperature involved (Volkovic, 1983). Detailed description of source profiles has been described elsewhere (Dubey, 2011).

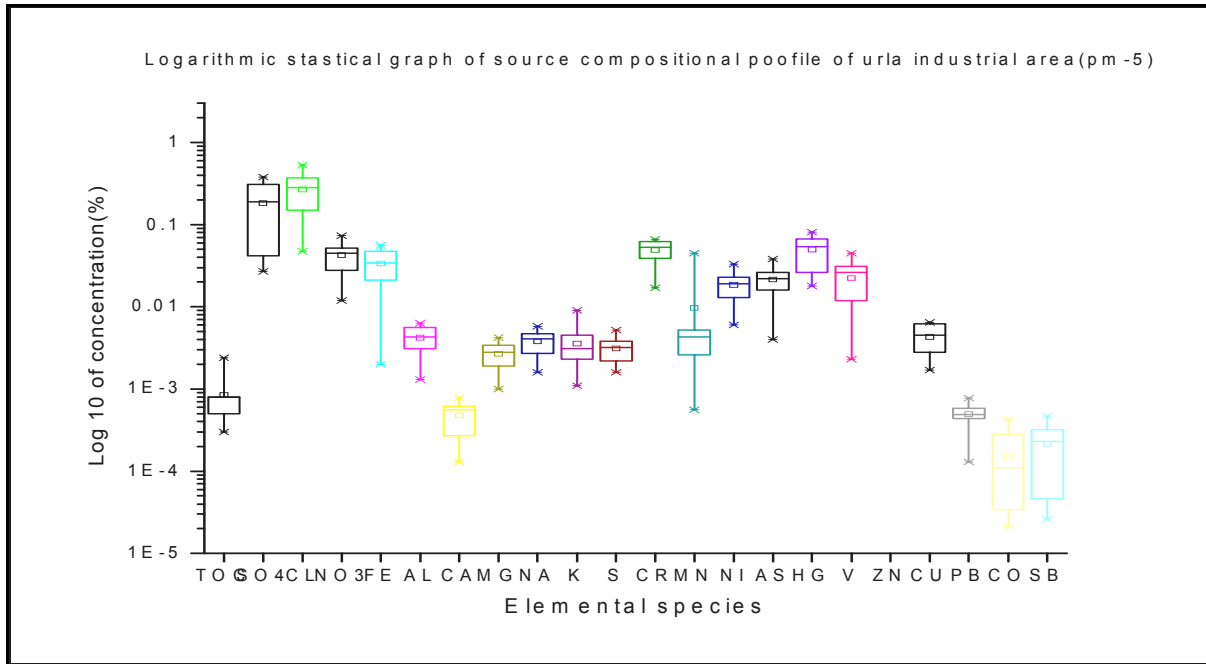
Source Apportionment Approach and Sensitivity Testing

Source apportionment was carried out in two steps: 1) Using linear regression analysis to evaluate the possible relationship between indoor-receptors and identified source-routes, and 2) using chemical mass balance receptor modeling (CMB 8) (USEPA) to quantify the relative source contributions. Geller *et al.* (2002) describes the intercept of regression line between dependent (Receptor RPM levels) and independent variables (Source RPM levels) has been identified as concentration of PM generated within receptor itself and not contributed by independent source. Similarly slope values of regression have been evaluated to be the decay constant of PM at dependent variable (receptor site). These factors (intercept and slope) were used to investigate probable transfer of source PM levels to a specific receptor site in earlier studies. A clear agreement between calculated and measured values has been obtained (Geller *et al.*, 2002; USEPA, 2003). The values of intercept and slope calculated in this study have been presented in Table 5. Watson *et al.* (1998) and Watson and Chow (2005) describe CMB modeling procedures and an application and validation protocol. The activities carried out to complete the protocol are described in this section.

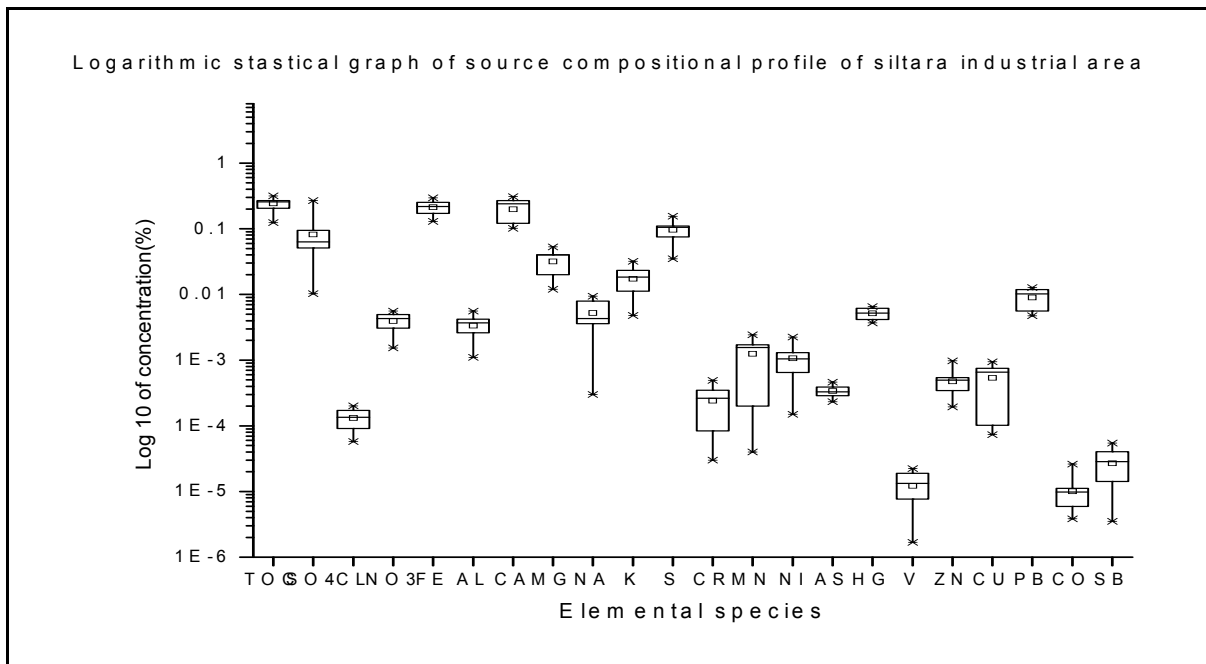
CMB Model Applicability: The requirements for CMB model applicability are as follows: (1) a sufficient number of receptor samples are taken with an accepted method to evaluate compliance with standards; (2) samples are analyzed for chemical species which are also present in source emissions; (3) potential source contributions have been identified and chemically characterized; (4) the number of non collinear source types is less than the number of measured species (Watson, 1984; Watson *et al.*, 1991;

Watson et al., 1997; Watson et al., 1998; Watson and Chow, 2005). All major source types in the study region have been identified, and profiles which represent their emissions have been assembled. Examination of the chemical profiles shows significant differences among profiles for major source types such as ferrous metallurgical process oriented industrial groups (Siltara Industrial area, S-6), chemical, fertilizers and steel rolling process based industrial groups (Urla Industrial area, S-5), road-traffic emissions, household fuel burning and

soils re-suspension. The number of species measured in source and receptor samples is greater than the number of non collinear sources. Initial tests with different combinations of profiles determine which profiles best explain the data at each site. Several initial CMB runs were performed at each site for indoor RPM, and the CMB performance measures were examined to determine how well the indoor concentrations were explained by the source contribution estimates. The best fit parameters have been presented in Table 6.

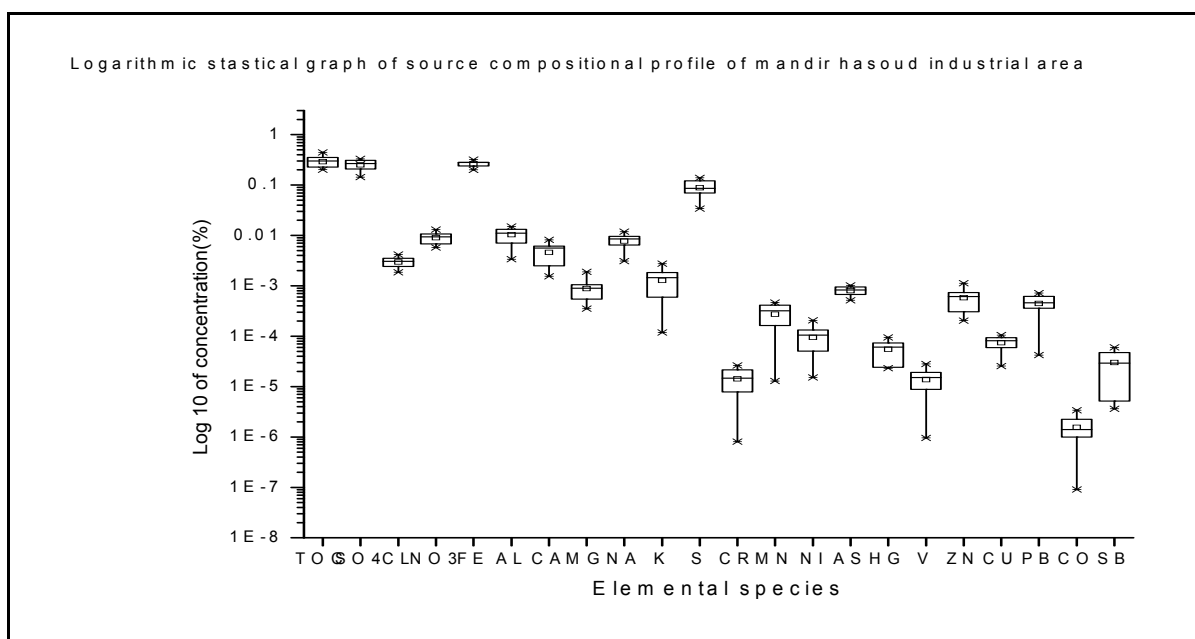


(A)

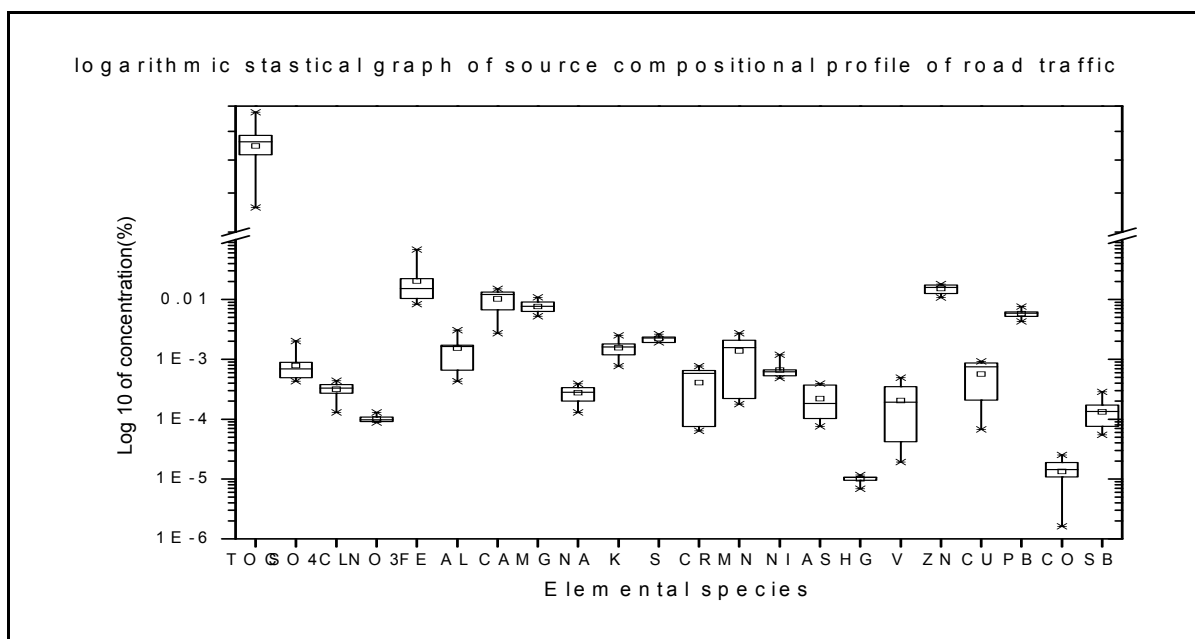


(B)

Fig. 3. Logarithmic statistical graph of source compositional profile of Downwind Ambient RPM near (A) Urla Industrial Area (S-5); (B) Siltara Industrial Area (S-6).



(A)



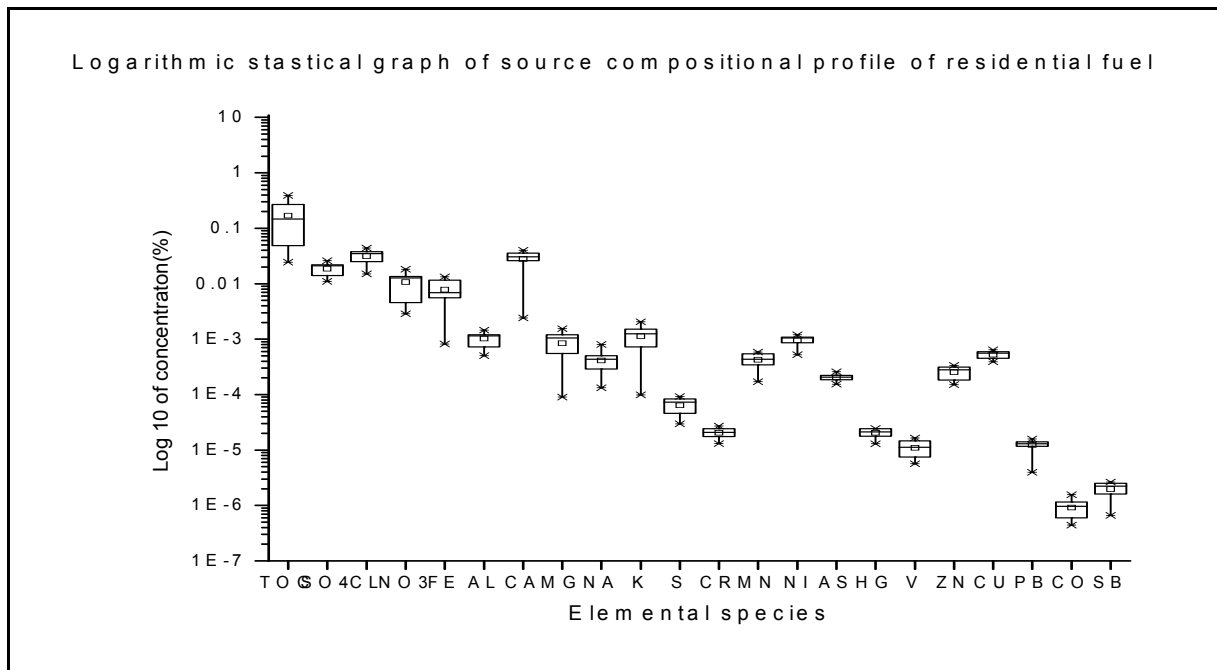
(B)

Fig. 3. Logarithmic statistical graph of source compositional profile of Downwind Ambient RPM near (A) Mandir Hasaud Industrial Area (S-7); (B) Road-Traffic (S-8).

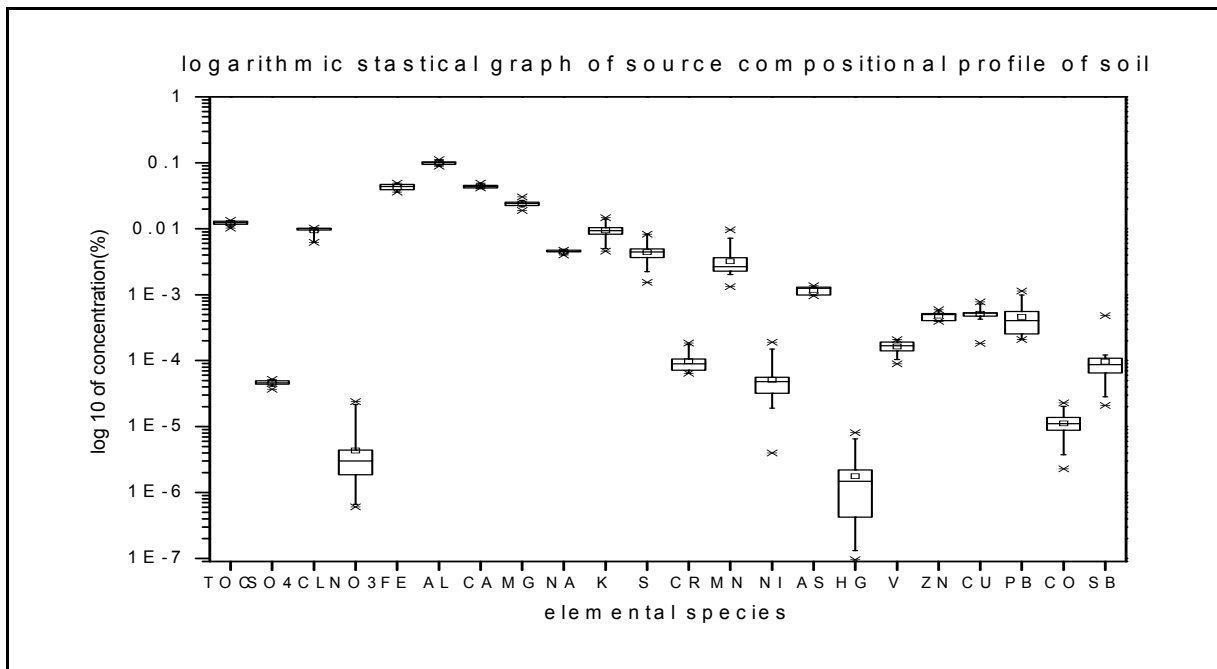
Source Contribution Estimates

Source apportionment results (Table 6 and Fig 4) comparing indoor RPM source contributions by linear regression and CMB. CMB results have shown clearer picture of source apportionment of indoor RPM with potential impact of S-6 in selected indoor-receptors. It has given contribution range from 25–39% of indoor RPM with exception at R-1 and R-2. S-5 has shown major contribution only at R-1 due to close proximity, which was not observed in regression analysis. Again, Mandir Hasaud (R-7) has shown more clarity of contribution with 51% in R-2 that explained

the temporary straight approach of S-7 to R-3 region. Road-traffic (S-8) has shown consistency across the indoor-receptor sites with contribution range of 16–31%. Soils (S-9) of the study region have not shown any contribution to indoor-receptors due the fact that road-traffic profile is developed as mixture of automobile emissions and roadside runoff. Introduction of HFB (S-10) in CMB has shown significant impact on source contribution pattern reported earlier by regression analysis where HFB was excluded. It has shown contribution range of 15–53% across the indoor-receptors with significant share of total source contribution and



(A)



(B)

Fig. 3. Logarithmic statistical graph of source compositional profile of RPM emitted from (A) Household fuel burning (S-10); (B) Soils (S-9).

Table 5. Regression analysis between Source-Receptor matrices using RPM data of residential-indoors and source-ambient.

Receptor \ Source	RPM (R^2) regression analysis											
	S-5			S-6			S-7			S-8		
	M	C	R^2	M	C	R^2	M	C	R^2	M	C	R^2
R-1	0.269	491.6	0.146	1.205	241.5	0.226	0.240	453.2	0.011	0.259	380	0.025
R-2	0.315	555.4	0.110	2.449	846.7	0.516	1.363	642.5	0.196	1.056	654.9	0.230
R-3	0.254	425.3	0.086	1.407	148.3	0.204	0.902	251	0.103	1.376	125.7	0.431
R-4	0.268	825.5	0.139	1.964	460.7	0.576	0.21	732.2	0.008	0.553	664.2	0.109

* M: Slope, C: Y intercept, R^2 : square of the correlation coefficient.

Table 6. CMB8 Good Fit parameter for Indoor RPM.

Subject	Date	R ²	χ ²	TSTAT					
				S-5	S-6	S-7	S-8	S-9	S-10
R-1	27/10/08	0.53	1.58	2.97	0.54	1.29	1.06	-1.05	0.46
R-2	02/11/08	0.08	11.1	0.92	1.21	-	0.81	-	2.13
R-3	07/11/08	0.28	9.15	-2.87	2.10	5.21	1.84	-2.0	2.56
R-4	12/11/08	0.31	3.54	2.38	1.26	-0.31	1.33	-1.14	0.86

TSTAT: T statistic, R²: R square, χ²: Chi Square.

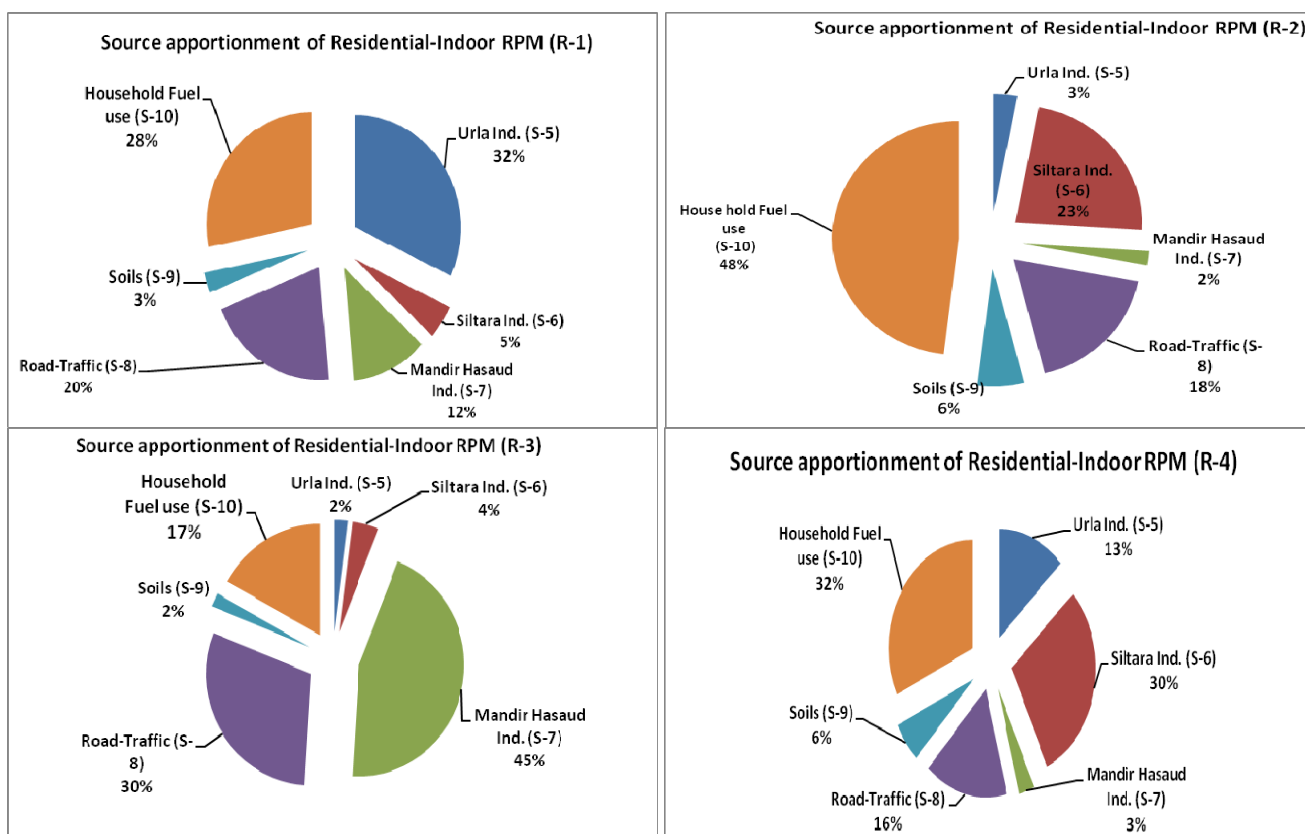


Fig. 4. Relative source contributions of RPM in environmentally defined indoor-receptors of Raipur, CG, India.

consequently decreases other outdoor sources except the dense plume approaches directly from S-7 to R-3. In case of R-4 indoor-receptor close to S-5, regression results have shown 53% contribution from S-5. HFB diminishes it to 39% with significant margin of 14%. As far as goodfit parameters is concern, S-5 has shown prescribed standards of TSTAT with R-1 and R-4, S-6 with R-3, S-7 with R-3, S-8 with R-3 and S-10 with R-2 and R-3. Occurrence of higher insignificant TSTAT in different combinations of source-receptors has attributed to need of detailed speciation of total carbon in indoor RPM.

CONCLUSION

The hypothesis of the study has been well explained experimentally. Results of regression analysis have explained the probable relations of selected source-receptors, but CMB results explained the detailed quantification of selected source contributions to RPM of specific indoor-receptor. The

conclusions drawn from regression analysis are: 1) probable relationship between selected source-routes and indoor-receptor with best correlation of Siltara Industrial area (S-6) to most of indoor-receptors, 2) Higher deviation pattern from mean RPM of Urala Industrial area (S-5) is mainly responsible for negative correlation with all indoor-receptors and 3) S-6 has been identified as main outdoor-source contributor (up to 50%) of indoor RPM in most of the sites. While CMB results are concluded as: 1) Outdoor source-routes have shown different approach among selected indoor-receptors compared to regression analysis, but again the Siltara Industrial area has been identified as main contributor to R-2 and R-4, while Urla (S-5) has shown major contribution to R-1 and Mandir Hasaud (S-7) to R-3, 2) Road-traffic has shown consistency in relative source contribution to indoor RPM across the sites, 3) HFB has shown significant variation in its relative contribution to indoor RPM (15–53%) across sites and suppressed the outdoor infiltration in selected indoor-receptors. 4) Soils have

not shown any contribution due to similarity with the source profiles of road-traffic emissions (S-8) and 5) In spite of taking 17 chemical species in CMB execution, the good fit parameters have not met the prescribed standards in a number of relationship cases. The probable reason behind this are: 1) Almost all source-routes carrying total carbon as major content, 2) Different combustion temperature involved across source types resulting in emission of varied proportion of speciated carbon species and 3) Hence lack of generation of speciated carbon species data might be the main reason and needed to be resolved. In spite of this, a preliminary picture of source apportionment of indoor RPM of Raipur, India has explained that industrial contribution to indoor RPM is mainly depend on wind pattern, whereas road-traffic emissions have shown consistency in relative source contribution with 1/3rd of total indoor RPM. Household fuel burning emissions (HFB) have shown varied impact on indoor RPM depending on the ventilation properties and fuel used in the houses.

ACKNOWLEDGEMENT

One of the authors (ND) is grateful to School of Studies in Chemistry, Pt. Ravishankar Shukla University, and Raipur for providing laboratory facilities and financial assistance. Author (SP) also grateful to Desert Research Institute, Reno, NV for providing library & computer facilities during Fulbright Fellowship.

REFERENCES

- Abt, E., Suh, H.H., Allen, G. and Koutrakis, P. (2000). Characterization of Indoor Particle Sources: A Study Conducted in the Metropolitan Boston Area. *Environ. Health Perspect.* 108: 35–44.
- Adgate, J.L., Willis, R.D., Buckley, T.J., Chow, J.C., Watson, J.G., Rhoads, G.G. and Lioy, P.J. (1998). Chemical Mass Balance Source Apportionment of Lead in House Dust. *Environ. Sci. Technol.* 32: 108–114.
- Balakrishna, G. and Pervez, S. (2009). Source Apportionment of Atmospheric Dust Fallout in an Urban-Industrial Environment in India. *Aerosol Air Qual. Res.* 9: 359–367.
- Basu, D., Saha, R., Ganguly, R. and Datta, A. (2008). Performance Improvement of LPG Cook Stoves through Burner and Nozzle Modifications. *J. Energy Inst.* 81: 218–225.
- Begum, B.A., Paul, S.K., Hossain, M.D., Biswas, S.K. and Hopke, P.K. (2009). Indoor Air Pollution From Particulate Matter Emissions in Different Households in Rural Areas of Bangladesh. *Build. Environ.* 44: 898–903.
- Bhargava, A., Khanna, R.N., Bhargava, S.K. and Kumar, S. (2004). Exposure Risk to Carcinogenic PAHs in Indoor-Air during Biomass Combustion Whilst Cooking in Rural India. *Atmos. Environ.* 38: 4761–4767.
- Borderieux, S., Wu, C.Y., Bonzongo, J.C. and Powers, K. (2004). Control of Elemental Mercury Vapor in Combustion Systems Using Fe₂O₃ Nanoparticles. *Aerosol Air Qual. Res.* 4: 74–90.
- Canepari, S., Cardarelli, E., Giuliano, A. and Pietrodangelo, A. (2006). Determination of Metals, Metalloids and Non-Volatile Ions in Airborne Particulate Matter by a New Two-Step Sequential Leaching Procedure Part A: Experimental Design and Optimisation.
- Cao, J.J., Lee, S.C., Chow, J.C., Cheng, Y., Ho, K.F., Fung, K.K., Liu, S.X. and Watson, J.G. (2005a). Indoor/Outdoor Relationships for PM_{2.5} and Associated Carbonaceous Pollutants at Residential Homes in Hong Kong - Case Study. *Indoor Air* 15: 197–204.
- Cao, J.J., Rong, B., Lee, S.C., Chow, J.C., Ho, K.F., Liu, S.X. and Zhu, C.S. (2005b). Composition of Indoor Aerosols at the Emperor Qin's Terra-Cotta Museum, Xi'an, China, during Summer 2004. *Particuology* 3: 170–175.
- Chow, J.C., Watson, J.G., Houck, J.E., Pritchett, L.C., Rogers, C.F., Frazier, C.A., Egami, R.T. and Ball, B.M. (1994). A Laboratory Resuspension Chamber to Measure Fugitive Dust Size Distributions and Chemical Compositions. *Atmos. Environ.* 28: 3463–3481.
- Cortez-Lugo, M., Moreno-Macias, H., Holguin-Molino, F., Chow, J.C., Watson, J.G., Gutierrez-Avedoy, V., Mandujano, F., Hernandez-Avila, M. and Romieu, I. (2008). Relationship between Indoor, Outdoor, and Personal Fine Particle Concentrations for Individuals with COPD and Predictors of Indoor-Outdoor Ratio in Mexico City. *J. Exposure Sci. Environ. Epidemiol.* 18: 109–115.
- Crist, K.C., Liu, B., Kim, M., Deshpande, S.R. and John, K. (2008). Characterization of Fine Particulate Matter in Ohio: Indoor, Outdoor, and Personal Exposures. *Environ. Res.* 106: 62–71.
- DeFries, R. and Pandey, D. (2010). Urbanization, the Energy Ladder and Forest Transitions in India's Emerging Economy. *Land Use Policy.* 27: 130–138.
- Dubey, N. (2011). Study of Particulate Source Apportionment at Classified Atmospheric Receptors in Selected Defined Urban Areas.
- Dubey, N. and Pervez, S. (2008). Investigation of Variation in Ambient PM₁₀ Levels within an Urban-Industrial Environment. *Aerosol Air Qual. Res.* 8: 54–64.
- Farsi, M., Filippini, M. and Pachauri, S. (2007). Fuel Choices in Urban Indian Households. *Environ. Dev. Econ.* 12: 757–774.
- Gadkari, N.M. and Pervez, S. (2007). Source Investigation of Personal Particulates in Relation to Identify Major Routes of Exposure among Urban Residentials. *Atmos. Environ.* 41: 7951–7963.
- Gadkari, N.M. and Pervez, S. (2008). Source Apportionment of Personal Exposure of Fine Particulates among School Communities in India. *Environ. Monit. Assess.* 142: 227–241.
- Ge, S., Xu, X., Chow, J.C., Watson, J.G., Sheng, Q., Liu, W., Bai, Z., Zhu, T. and Zhang, J. (2004). Emissions of Air Pollutants from Household Stoves: Honeycomb Coal versus Coal Cake. *Environ. Sci. Technol.* 38: 4612–4618.
- Geller, M.D., Chang, M.H., Sioutas, C., Ostro, B.D. and Lipssett, M.J. (2002). Indoor/Outdoor Relationship and Chemical Composition of Fine and Coarse Particles in the Southern California Deserts. *Atmos. Environ.* 36: 1099–1110.
- Gilbert, R.O. (1987). *Statistical Methods for Environmental*

- Pollution Monitoring*, John Wiley, New York.
- Hidy, G.M., Lachenmyer, C., Chow, J.C. and Watson, J.G. (2000). Urban Outdoor-Indoor PM_{2.5} Concentrations and Personal Exposure in the Deep South: Part II. Inorganic Chemistry. *Aerosol Sci. Technol.* 33: 357–375.
- Hu, T., Li, X., Dong, J., Rong, B., Shen, Z., Cao, J.J., Lee, S.C., Chow, J.C. and Watson, J.G. (2006). Morphology and Elemental Composition of Dustfall Particles Inside Emperor Qin's Terra-Cotta Warriors and Horses Museum. *Particuology* 4: 346–351.
- Hu, T.F., Lee, S.C., Cao, J.J., Chow, J.C., Watson, J.G., Ho, K.F., Ho, W.K., Rong, B. and An, Z.S. (2009). Characterization of Winter Airborne Particles at Emperor Qin's Terra-Cotta Museum, China. *Sci. Total Environ.* 407: 5319–5327.
- Joon, V., Chandra, A. and Bhattacharya, M. (2009). Household Energy Consumption Pattern and Socio-Cultural Dimensions Associated With It: A Case Study of Rural Haryana, India. *Biomass Bioenergy* 33: 1509–1512.
- Kanishtha, T., Banerjee, R. and Venkataraman, C. (2006). Effect of Particle Emissions from Biofuel Combustion on Surface Activity of Model and Therapeutic Pulmonary Surfactants. *Environ. Toxicol. Pharmacol.* 22: 325–333.
- Kopperud, R.J., Ferro, A.R. and Hildemann, L.M. (2004). Outdoor Versus Indoor Contributions to Indoor Particulate Matter (PM) Determined by Mass Balance Methods. *J. Air Waste Manage. Assoc.* 54: 1188–1196.
- Koshle, A., Pervez, Y.F., Tiwari, R.P. and Pervez, S. (2008). Environmental Pathways and Distribution Pattern of Total Mercury among Soils and Groundwater Matrices around an Integrated Steel Plant in India. *J. Sci. Ind. Res.* 67: 523–530.
- Kulshrestha, A., Bisht, D.S., Masih, J., Massey, D., Tiwari, S. and Taneja, A. (2009). Chemical Characterization of Water-Soluble Aerosols in Different Residential Environments of Semi Aridregion of India. *J. Atmos. Chem.* 62: 121–138.
- Larson, T., Gould, T., Simpson, C., Liu, L.J.S., Claiborn, C. and Lewtas, J. (2004). Source Apportionment of Indoor, Outdoor, and Personal PM_{2.5} in Seattle, WA, Using Positive Matrix Factorization. *J. Air Waste Manage. Assoc.* 54: 1175–1187.
- Massey, D., Masih, J., Kulshrestha, A., Habil, M. and Taneja, A. (2009). Indoor/Outdoor Relationship of Fine Particles Less Than 2.5 μm (PM_{2.5}) in Residential Homes Locations in Central Indian Region. *Build. Environ.* 44: 2037–2045.
- Mondal, N.K., Mukherjee, B., Das, D. and Ray, M.R. (2010). Micronucleus Formation, DNA Damage and Repair in Premenopausal Women Chronically Exposed to High Level of Indoor Air Pollution From Biomass Fuel Use in Rural India. *Mutat. Res.-Genetic Toxicol. Environ. Mutagen.* 697: 47–54.
- Montaser, A. and Golightly, D.W. (1987). *Inductive Coupled Plasma in Analytical Atomic Spectrometry*, VCH, New York.
- Padhi, B.K. and Padhy, P.K. (2008). Domestic Fuels, Indoor Air Pollution, and Children's Health: The Case of Rural India. *Ann. N.Y. Acad. Sci.* 1140: 209–217.
- Pohekar, S.D., Kumar, D. and Ramachandran, M. (2005). Dissemination of Cooking Energy Alternatives in India - A Review. *Renewable Sustainable Energy Rev.* 9: 379–393.
- Pokhrel, A.K., Smith, K.R., Khalakdina, A., Deuja, A. and Bates, M.N. (2005). Case-Control Study of Indoor Cooking Smoke Exposure and Cataract in Nepal and India. *Int. J. Epidemiol.* 34: 702–708.
- Rai, K., Sarkar, A.K. and Mitra, A.P. (2002). In *Aerosol Remote Sensing in Global Change and Atmospheric Pollution*, IASTA Bulletin, Indian Aerosol Science and Technology Association, Thiruvananthapuram, p. 155.
- Raiyani, C.V., Shah, S.H., Desai, N.M., Kenkaiah, K., Patel, J.S., Parikh, D.J. and Kashyap, S.K. (1993). Characterization and Problems of Indoor Pollution Due to Cooking Stove Smoke. *Atmos. Environ.* 27: 1643–1655.
- Sexton, K. and Hayward, S.B. (1987). Source Apportionment of Indoor Air Pollution. *Atmos. Environ.* 21: 407–418.
- Thakur, M., Deb, M.K., Imai, S., Suzuki, Y. and Ueki, K. (2004). Load of Heavy Metals in the Airborne Dust Particulates of an Urban City of Central India. *Environ. Monit. Assess.* 95: 257–268.
- Tursic, J., Radic, H., Kovacevic, M., and Veber, M. (2008). Determination of Selected Trace Elements in Airborne Aerosol Particles Using Different Sample Preparation. *Arch. Ind. Hyg. Toxicol.* 59: 111–116.
- USEPA (2003). Fourth External Review Draft of Air Quality Criteria for Particulate Matter.
- Verma, S.K., Deb, M.K., Suzuki, Y. and Tsai, Y.I. (2010). Ion Chemistry and Source Identification of Coarse and Fine Aerosols in an Urban Area of Eastern Central India. *Atmos. Res.* 95: 65–76.
- Volkovic, V. (1983). *Trace Elements in Coal*, CRC, Florida.
- Watson, J.G. (1984). Overview of Receptor Model Principles. *J. Air Pollut. Contr. Assoc.* 34: 619–623.
- Watson, J.G. and Chow, J.C. (2005). In *Air Quality Modeling -Theories, Methodologies, Computational Techniques, and Available Databases and Software*. Vol. II - Advanced Topics, Zannetti, P. (Ed.), Air and Waste Management Association and the EnviroComp Institute, Pittsburgh, PA, p. 455.
- Watson, J.G., Chow, J.C. and Pace, T.G. (1991). In *Receptor Modeling for Air Quality Management*, Hopke, P.K. (Ed.), Elsevier Press, New York, NY, p. 83.
- Watson, J.G., Robinson, N.F., Lewis, C.W., Coulter, C.T., Chow, J.C., Fujita, E.M., Conner, T.L. and Pace, T.G. (1998). CMB8 Application and Validation Protocol for PM_{2.5} and VOCs, Desert Research Institute Document, No. 1808.2D1.
- Watson, J.G., Robinson, N.F., Lewis, C.W., Coulter, C.T., Chow, J.C., Fujita, E.M., Lowenthal, D.H., Conner, T.L., Henry, R.C. and Willis, R.D. (1997). Chemical Mass Balance Receptor Model Version 8 (CMB) User's Manual. Desert Research Institute Document, No. 1808.1D1.

Received for review, August 10, 2011

Accepted, November 20, 2011