Source Apportionment of PM2.5 Using a CMB Model for a Centrally Located Indian City

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ABSTRACT

Samples of PM2.5 were collected sequentially for 24 hours during the last week of September to mid February 2009–10 at three locations representing residential (R), commercial (C) and industrial (I) sites in Nagpur city to determine their chemical composition and estimations of the sources contributing to them. Two receptor models were used for the source apportionment viz. enrichment factors (EF) to differentiate crustal and non-crustal sources, whereas chemical mass balance (CMB 8.2) was used to identify and quantify the major sources contributing to PM2.5.

The ambient mass concentrations and chemical compositions of PM2.5 with respect to ionic species (Na^+, NH_4^+, K^+, Ca^{2+}, F^-, Cl^-, NO_3^- and SO_4^{2-}); carbonaceous species (organic and elemental carbon) and trace metals (Al, Ba, Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Si and Zn) were determined. The most abundant chemical species were OC, EC, SO_4^{2-}, NO_3^-, NH_4^+, K^+ and trace metals (Al, Fe, Si, Mg, and Cu) at all the sites.

Findings of EF showed the anthropogenic origin of Cd, Ni, Pb, Cu, Fe and Zn, whereas Ba, Cr, Mg, Mn, and Si were contributed from crustal sources. On the other hand, results of CMB using source profiles developed in India for non-vehicular and vehicular sources revealed that vehicular emissions were major contributing sources 57, 62 and 65%; followed by secondary inorganic aerosol 16, 12, 16%; biomass burning 15, 11, 9% and then by re-suspended dust 6,10, 7% at R, C and I sites, respectively. This study showed that while the sources at all three sites were mostly consistent, the percent contributions of these varied among the sites as per the intensity of ongoing activities at the receptor sites.

Keywords: PM2.5; Metals; Anions-Cations; OC-EC; Source apportionment; CMB.

INTRODUCTION

Nagpur is a centrally located fast growing metropolis city in India. It is the second capital of Maharashtra having an approximate population of 2.5 million people. This city has railway loco shed, international airport, major industries like automobiles, chemicals, electronics, agro based industries, thermal power plants (12–15 km away from the city) and more than 1000 small and medium scale industries.

Presently, only PM10 (particles having aerodynamic diameter dp < 10 μm) are being monitored in the Nagpur city. As per the NEERI report (2010), annual levels of PM10 are exceeding the limits i.e., (annual- 60 μg/m³) given by Central Pollution Control Board (CPCB). One of the studies carried out for Nagpur estimated that approximately 5.03 Mg/km² emission of particulate matter (PM) was from

5,43,322 vehicles (Ramchandra and Swetmala, 2009). During our study period, the number of vehicles were almost doubled i.e., (11, 10, 225) with annual vehicular growth rate of 10.2% and out of total vehicles, nearly 84% two wheelers were registered in Nagpur. Furthermore, upcoming mega projects like multi-modal international cargo hub and airport at Nagpur (MIHAN) would enhance the economic growth rate and population of Nagpur at a faster pace (Meshram, 2011). With this, dramatic increase in the number and density of vehicles in the city, the pollution of particulates would increase further to substantial amount. As far as domestic sector is concerned, LPG is mostly used in residential homes. But in hotels and restaurants along with LPG, either coal or wood is used. Very often, economically weaker sections use wood for cooking and water heating purpose. In addition to this, people burn biomass outside the houses to get the warmth during winter. Biomass is also burned to clean the surrounding areas by local residents or sometimes by the Nagpur Municipality workers. Thus, the local activities are also responsible for increasing the levels of particulate matter. In a tropical city like Nagpur, natural ventilation is preferred which results in entering the pollution

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from outside into the residential buildings. Thus, people get exposed to this kind of pollution every day. Recent findings have shown that $PM_{2.5}$ (particles having aerodynamic diameter $dp < 2.5$ μm) comprises several primary and secondary components including ionic, mutagenic/carcinogenic organics and toxic trace metals and pose threat to human health (Schwartz, *et al.*, 1996; Ostro, *et al.*, 2006). They are also responsible for reducing the visibility (Sisler and Malm, 2000) and affecting the global climate (Charlson *et al.*, 1992; Wexler and Ge, 1998). In view of this, CPCB has introduced a standard for $PM_{2.5}$ applicable to mixed (residential, industrial and economically sensitive) areas. Therefore, it is imperative to identify and quantify major possible sources contributing to $PM_{2.5}$ so that effective control management plan can be developed to reduce the levels of $PM_{2.5}$ at the initial stages of development of the city.

Better estimation of source apportionment of PM can be done by using receptor models. Several receptor models such as enrichment factors (EFs), chemical mass balance (CMB), eigenvector analysis (also termed principal component analysis (PCA) and empirical orthogonal functions (EOF), multiple linear regression, neural networks, edge detection, cluster analysis, Fourier Transform time series and a number of other multivariate data analysis methods are used for identifying and quantifying the sources of air pollutants at a receptor location.

Out of these receptor models, (CMB 8.2, USEPA, 2004) is used in this study as it estimates the contribution of sources by determining the best-fit combination of chemical profiles of emission sources and chemical composition of ambient particulates (Watson *et al.*, 1991; Watson *et al.*, 1994). One more advantage of this model is that only few

measurements are required and even a single ambient sample analysis is possible. Source profiles required by this model are available in USEPA speciate 3.2 data base but as per the findings of Sharma and Patil (1993), these source profiles were not suitable for highly polluted Indian cities. Therefore, source profiles for non-vehicular sources (IITB Report, 2008, http://www.cpcb.nic.in/source_emiss ion_%20profiles_NVS_volume%20one.pdf) and vehicular sources (ARAI Report, 2008; www.cpcb.nic.in/source_pr ofile_vehicles.pdf) were developed in India. These source profiles are used in CMB along with ammonium sulfate, and ammonium nitrate profiles based on the calculated mass balance for the source apportionment of $PM_{2.5}$ whereas EFs is used to identify the nature of the sources at the receptor sites.

METHODOLOGY

Sampling Sites

Samples of $PM_{2.5}$ were collected at residential (R), commercial (C) sites (both at the height of 5 m) and industrial site (I) (at the height of 7 m) as shown in Fig. 1. Sequential sampling was carried out and twenty eight samples were collected during 25th September to 5th November 2009, 11th December 2009 to 12th January 2010 and 13th January to 12th February 2010 at R, C and I sites respectively.

As the city is expanding, no demarcation can be made in the respective activities. All the selected sites are mainly residential locations in R, C and I areas. R site represent residential cum commercial area. It is densely populated and congested area with narrow roads having high density of two, four wheelers. Commercial activities include gold

🔴 Residential site   🟢 Commercial site   🔵 Industrial site

Fig .1. Locations of sampling sites.

smelting shops, cloth and grocery shops. At R site, frequent traffic jams due to narrow congested roads and occasional biomass burning along the road side are observed. Major shops are located around the site C with heavy traffic flow of two, three and four wheelers along with other commercial vehicles. During the study period, traffic density around this site was about 30,000 vehicles per day. City bus stand is also located in the same area. Biomass burning was going on during the sampling period, on the agricultural land situated around 0.7 km downwind to the sampling site. At I site, sampling was performed at the roof of a marriage hall where cooking is done using charcoal or mostly with wood in tandoor (it is a cylindrical shaped heater made from clay where heat is generated by a wood or charcoal fire for cooking and baking). Industrial activity was located 2.5 km away from sampling site (I) and to the upwind direction. Heavy traffic of trucks, two, three and four wheelers were observed around this site during the study period. The major traffic that was observed of the heavy duty trucks during the night time with a speed of 35–45 km/hr. In addition to this; occasional roadside biomass burning was going on during the sampling period.

Sample Collection

Partisol Model 2300, 4-channel speciation sampler, (Thermo Fisher Scientific Inc., USA) was used to collect samples through an inlet at a flow rate of 16.7 LPM. A flow device (Streamline-PRO flow calibrator) supplied with the instrument was used to calibrate the flow rate prior to and after the sampling was over. The flow rate was within the $16.7 \pm 0.2\%$ LPM. $PM_{2.5}$ samples were collected on different filter media such as teflon (2 μm PTFE Whatman® Teflon, 7592–104) for metals, nylon (2 μm, Nylasorb™ pall flex, 66509) for ions and quartz micro fiber (Pall flex Tissue quartz, 7202, 2500QAT-UP) filters for OC and EC. Simultaneously, meteorological parameters such as wind speed (WS), wind direction (WD), temperature (T), relative humidity (RH) were measured at the sampling sites using weather station (make LSI-LASTEM, s.r.l., Italy).

Gravimetric and Chemical Analyses

Before and after the sampling procedure, filters were kept for 48 h in a desiccators in an environmentally conditioned room with a RH of $45 \pm 4\%$ and a temperature of $25 \pm 3°C$ before being weighed by a microbalance (Mettler AE 163) having precision of ± 10 μg. $PM_{2.5}$ concentrations were determined by gravimetric analysis using the weight difference of the filters before and after the sampling.

The quartz filters were preheated in an electric furnace at 800°C for about three hours in order to remove residues in the filters prior to use for sampling. As per USEPA protocol (1998), laboratory blanks (n = 7) and field blanks (n = 7) were collected as a part of QA/QC.

Analysis of Trace Metals

Trace metals (Al, Ba, Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Si and Zn) were determined by inductively coupled plasma optical emission spectrometry, (ICP-OES, Optima 4100 DV, Perkin Elmer, USA) following USEPA compendium

method (IO-3.1 and 3.4). $PM_{2.5}$ samples were digested with concentrated nitric acid (Merck, H17H570629) in a Teflon vessel in a microwave digestion chamber (ETHOS make-milestone, Italy) for twenty minutes and then filtered through Whatman 42 (Ashless filter papers 125 mm, Cat No. 1442 125) filters into properly cleaned volumetric flask. The concentration of metals in blank teflon filters processed similarly as that of samples (n = 7) were in the range of 0.0023 to 0.57 mg/L. These values were subtracted from the sample values to correct the final data. The Limit of Detection (LOD) for metals were estimated as the concentration corresponding to three times the standard deviation of the blank signals (3σ) obtained from a set of reagent blanks (n = 7). LOD for Al, Ba, Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Si and Zn were 39, 2.8, 6.1, 3.9, 7.8, 17.4, 6.0, 3.0, 3.3, 4.5, 25.7, 35.5 ng/m³ respectively. Similarly, limit of quantification (LOQ) were estimated as 10 times the standard deviation of the blank signals obtained from a set of reagent blanks (n = 7). LOQ for Al, Ba, Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Si and Zn were found to be 120, 6.5, 18.1, 13.0, 23.0, 55.0, 20.0, 10.0, 10.6, 14.9, 85.0 and 120.0 ng/m³ respectively. Blank filters were spiked with a known amount of metals concentration and processed as per the protocol to calculate recovery efficiencies. Range of recovery efficiencies varied from 84.0 to 92.0% for the metals.

Analysis of Anions/Cations

Water soluble ions, four anions (F^-, Cl^-, NO_3^-, SO_4^{2-}) and four cations (Na^+, NH_4^+, K^+ and Ca^{2+}) were determined in aqueous extracts (ultrapure water, resistivity of 18 MΩ) of the filters. Samples were processed according to the standard method (SOP MLD 064, CARB). Exposed nylon filter was placed in a vial with 25 mL of ultra-pure water and sonicated for 60 min. F^-, Cl^-, SO_4^{2-}, NO_3^-, $NH4^+$, Na^+, Ca^{2+} and K^+ were determined by ion chromatograph (Dionex, DX 100) through separation of anionic analytes on a guard column preceded by a Dionex Ion Pac AS11 analytical column and separation of cationic analytes on a guard column preceded by a Dionex Ion Pac CS11analytical column. For anion separation, eluent was 5 mM NaOH having flow rate of 1.0 mL/min while for cation separation, eluent used was 6 mM methyl sulphonic acid (Merck) with a flow rate of 1.0 mL/min. Calibrations were performed using certified standards (Accu individual standard for anions and mixed standard for of IC-MCA), which were diluted to required concentration range (1, 5, 10 and 20 μg/L). Data acquisition, peak integration and calibration curves were performed by Chrome 2004 software package. LOQ for F^-, Cl^-, SO_4^{2-}, NO_3^-, NH_4^+, Na^+, Ca^{2+} and K^+ were 0.10, 0.11, 0.18, 0.12, 0.13, 0.33, 0.30 and 0.18 μg/m³ respectively.

Analysis of OC/EC

Elemental carbon (EC) and organic carbon (OC) were analyzed using DRI Model 2001 Thermal and Optical Carbon Analyzer, based on the preferential oxidation of OC and EC compounds at different temperatures (Chow *et al.*, 2004), with the IMPROVE thermal/optical reflectance (TOR) protocol (Chow *et al.* 1993). LOQ for OC and EC were 0.33 and 0.25 μg/m³ respectively.

RESULTS AND DISCUSSION

PM$_{2.5}$ Levels

Average PM$_{2.5}$ mass concentrations measured at R, C and I sites are plotted in Fig. 2. Temp., RH, WS and WD recorded at three different sites on hourly basis during the sampling were averaged over the sampler 24-hr operational time.

Concentrations of PM$_{2.5}$ were in the range of 31.5–129.5 μg/m^3 (mean 67.1 ± 22.1 μg/m^3) at R site. At this site, temperature, RH, WS were in the range of 25.2–31.0°C (mean 27.7°C), 44–55% (mean 47.5%) and 0.2–0.9 km/h (mean 0.45km/h) respectively. WD dominated from E and NNE with 37% calm conditions.

At C site, PM$_{2.5}$ were in the range of 56.2–161.7 μg/m^3 with mean 96.4 ± 21.6 μg/m^3. Here, temperature RH, WS were in the range of 19.0–24.1°C (mean 21.0°C), 56–68% (mean 62.7%), 0.2–0.93 km/h (mean 0.47 km/h) respectively. WD dominated from E and NE, with 61% calm conditions. At I site, the levels of PM$_{2.5}$ were in the range 54.7–118.4 μg/m^3 (mean 85.3 ± 16.2 μg/m^3) whereas temperature was in the range of 18.0–25.0°C (mean 21.9°C), RH was in range of 45–57% (mean 50.0%). WS was ranged between 0.4 and 0.8 km/h (mean 0.52 km/h). WD in winter season dominated from E and NNE with 64% calm conditions.

Average PM$_{2.5}$ concentrations at all the sites were exceeded by 1.1 to 1.6 times to that of daily average (60 μg/m^3) of Indian National Ambient Air Quality Standard (NAAQS) guidelines promulgated by CPCB. Results showed that the average concentration at C site was higher than site I followed by site R. The possible reason for high concentration at C site might be due to air stagnation and low mixing height with slow traffic flow because of crowded area and narrow roads. Comparatively lower PM$_{2.5}$ levels were observed at I site which may be due to less crowded traffic due to wider roads and comparatively higher sampling height than at site C. Calm conditions with low wind speed usually favor to build up high concentration of PM$_{2.5}$ in the atmosphere. At C and I sites, calm conditions (61–64%) were prevailed

during the study period showing higher concentration than the site R where calm conditions observed were only (37%). Although sporadic information is available on PM$_{2.5}$ in India, the levels measured in this study were lower than the concentrations observed for PM$_{2.5}$ in some cities in India, Mumbai (108.7 μg/m^3), Delhi (268 μg/m^3) and Kanpur (225.2 μg/m^3) (CPCB, 2010) whereas were comparable to the concentrations observed at Pune (72.5 μg/m^3), Chennai (63.2 μg/m^3), and Bangalore (39.2 μg/m^3) (CPCB, 2010). The levels of PM$_{2.5}$ thus may vary according to the nature of sources available in the vicinity of sampling sites, meteorological and geological conditions and intensity of ongoing local activities.

Characterization of PM$_{2.5}$
Levels of Trace Metals

Concentration of metals (ng/m^3) observed at R, C and I for Al, Fe, Mg, Cu, Si, and Zn are shown in Fig. 3(a) & for Ba, Cd, Cr, Mn, Ni, Pb in Fig. 3(b) respectively. Out of total metallic composition observed at site R, contribution of Al was 36% followed by Mg (20%), Fe (13%), Si (11%) and Cu and Zn (both 6%). The contributions of other metals (Ba, Pb, Mn and Cr) were ranged between 1 and 3% to the total metal concentration. At site C, the contribution of Si was 41% followed by Al (24%), Mg (9%), Fe and Cu (both 7%) and Zn (6%) to the total metal concentration whereas the contribution of other metals (Ba, Pb, Mn, Cd, and Cr) were in the range of 1–3% to the total metals. At site I, contribution of Al was 44.4% followed by Cu (14.6%), Si (11.9%), Zn (8.89%), Mg (8.86%) and Fe (6.34%). The contribution of other metals (Ba, Pb, Mn, Cd) were in the range of 0.3–2% to the total metal concentration.

Among all metals, the concentration of Al (a crustal element) was higher at site R and site I as compared to commercial site but the concentration of Si, one of the major component of Earth"s crust was higher at commercial site by 3.5 to 4.2 times as compared to site R and I. This may be due to the open agricultural land which belongs to the Agriculture College. Zn is a reliable tracer of unleaded
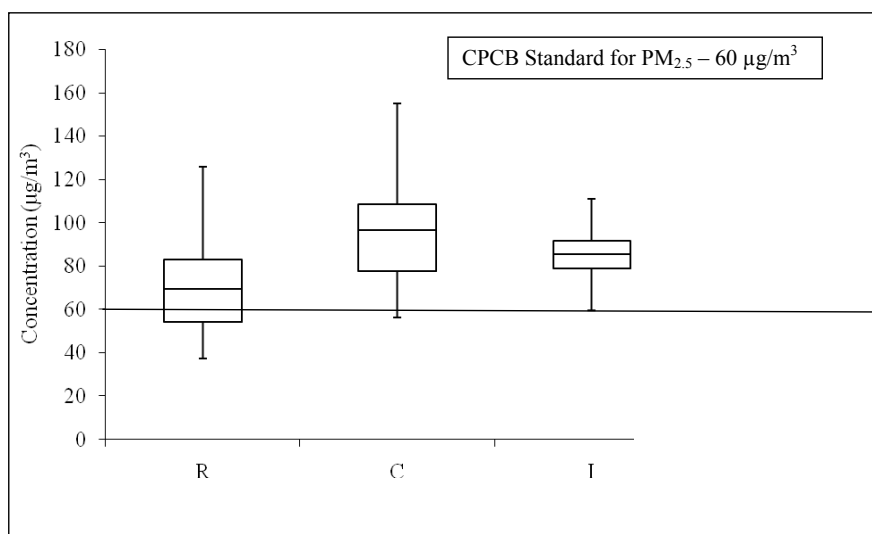

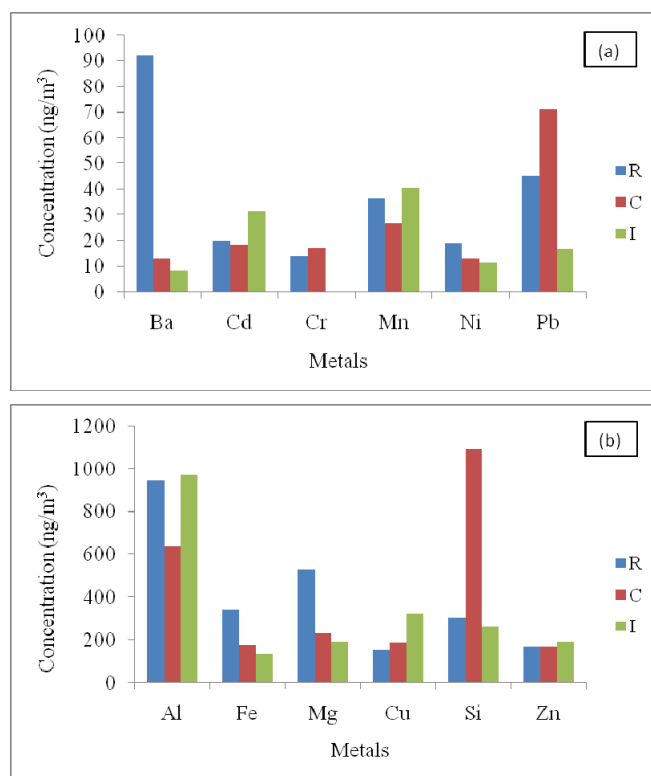**Fig. 2.** Average mass concentration of PM$_{2.5}$ (μg/m^3).

Fig. 3. Average mass concentration of metals in PM$_{2.5}$.

fuel and diesel oil powered motor vehicle emissions (Monaci *et al.*, 2000). It also emits from wood combustion (Mohn *et al.*, 2002). The concentration of Zn was nearly same for site R and C but slightly higher at site I indicating comparatively higher sources of Zn at the site. In urban areas, Cu is associated with road traffic (diesel engines and wearing of brakes). Concentration of Cu was higher at site I followed by site C and then by site R. Zn and Cu were higher at I may be due to diesel driven vehicles. The concentration of Pb was higher at site C followed by site R which may be due to the vehicular (Westerlunde, 2001) and biomass burning activities (Mohn et al., 2002). It was found that the concentration of Fe, Mg (fossil fuel combustion), Ba (road dust) and Ni (vehicular traffic) were higher at R site than the other two sites. The high concentration of Ba observed at R site was due to the occasional firecracker activity during the sampling period. The concentration of Cu, Cd and Mn were higher at industrial site which may be due to heavy traffic of vehicles and to some extent from resuspended dust. Concentrations of Pb (1.0 µg/m^3 for 24 hours) and Ni (20 ng/m^3 annually) were well below the CPCB prescribed standards at all the three sites.

The average metal concentrations observed in this study are compared with other studies carried out at different urban areas in India as well as in other Asian cities. Results tabulated in Table 1 show that levels of most of the metals (Al, Ba, Cd, Cr, Cu, Zn) are higher than the values reported at Tianjin, Beijing and Dhaka (Salem *et al.*, 2008; Gu *et al.*, 2011; Yang *et al.*, 2011) but are lower as compared to Kanpur city may be owing to its high industrial and vehicular growth.

Enrichment Factor

EF was applied to the quantified metals in PM$_{2.5}$ to evaluate the influence of crustal and anthropogenic sources at each receptor sites. In this study, Al was used as the reference element with upper continental crustal composition given by Mason (1966).

The crustal EF$_c$ was estimated using the following equation.

$$EF_c = \frac{\left(C_x/Al\right)_{PM_{2.5}}}{\left(C_x/Al\right)_{crust}} \tag{1}$$

where $(C_x/Al)_{PM2.5}$ = concentration ratio of metal to Al in PM$_{2.5}$, and $(C_x/Al)_{crust}$ = Concentration of metal to Al in crustal matter. If EF ≤ 10, it is considered that metals in PM$_{2.5}$ have a significant crustal contribution, and hence termed as the non-enriched elements. The EF > 10 indicates that metals have an important proportion of non-crustal sources and hence termed as the enriched elements (Zhang *et al.*, 2002). In this study, we used average earth crustal compositions as references in calculating EFc. The results of EF at three sites are tabulated in Table 2.

EF values for Cd were higher, followed by Ni, Pb, Cu, Fe and Zn and their EFs were greater than 10 indicating PM$_{2.5}$ have higher contribution from anthropogenic sources (Wu *et al.*, 2007). In this study area, enrichment of these metals at all sites could be attributed to local vehicular, biomass burning and other combustion activities. While the other metals such as Ba, Cr, Mg, Mn and Si showed EF less than 10, which suggests their crustal origin nature at all the sites.

Table 1. Comparison of metals of present study with major cities in India and other Asian countries (ng/m³).

Cities	Authors	Sampling Duration	Al	Ba	Cd
Nagpur	Present study	Sept–Feb 2009	852.5	37.8	18.0
Bay of Bangal	Shreenivas *et al.*,	Dec–Jan 2008–09	349	-	1.0
Kanpur	Behera and Sharma	Dec–Jan 2007–08	62–214	-	-
Hyderabad	Gummeneni *et al.*, 2011	Dec–Feb 2004–05	-	-	7.7
Tianjin, China	Gu *et al.*, 2011	6–21 Jan 2008	69.8	15.2	0.4
Dhaka, Bangladesh	Salem *et al.*, 2008	Jan and April 2006	-	-	6.3
Beijing, China	Yang *et al.*, 2011	Feb–April 2005–06	790	21	5.0
Kwun Tong, Hongkong	Ho *et al.*, 2006	Nov 2000–Feb 2001	161.9	-	-

Cr	Cu	Fe	Mg	Mn	Ni	Pb	Si	Zn
12.1	221.6	218.9	318.2	34.6	14.5	42.7	554.6	178.0
-	-	144	56	-	-	21	-	-
52–246	627–1453	146–549	503–1417	54–239	5–29	376–1568	1599–3420	295–601
6.1	2.0	502.8	-	46.8	12.8	418.5	-	200.5
-	38.9	145.1	819.5	32.3	143.2	231.1	1270.1	339.4
-	13.0	94.0	-	--		204	-	381.0
5.0	7.0	1130	290	90	20	240	1790	530.0
-	17.32	253.37	105.7	-	-	-	-	286.56

Table 2. Enrichment factors of metals in $PM_{2.5}$

Metals	Enrichment Factor		
	R	C	I
Ba	3.37	3.657	2.80
Cr	0.20	0.313	0.205
Cd	9087.83	14543.31	7083.85
Cu	188.64	343.24	382.46
Fe	146.11	146.98	80.76
Mg	2.16	1.06	0.55
Mn	3.09	3.42	3.52
Ni	200.39	113.33	161.78
Pb	160.43	566.66	61.18
Si	0.094	0.50	0.078
Zn	109.63	163.95	122.74

Concentration of Anions and Cations

Ionic abundances of inorganic compounds followed the trends $SO_4^{2-} > NO_3^- > NH_4^+ > K^+ > Cl^- > Ca^{2+} > F^- Na^+$ at all the sites as shown in Fig. 4. Among all the ions, SO_4^{2-} was the most abundant species (7.96, 8.60 and 10.07 µg/m³) followed by NO_3^- (6.43, 7.70 and 7.06 µg/m³), NH_4^+ (7.11, 5.40 and 7.20 µg/m³), K^+ (3.29, 2.82 and 3.64 µg/m³), Cl^- (1.09, 1.47 and 0.47 µg/m³), F^- (0.53, 0.59, 0.50 µg/m³), Ca^{2+} (0.55, 0.93, 0.64 µg/ m³) and Na^+ (0.88, 0.81, 0.28 µg/m³) at R, C and I sites respectively. The concentrations of Ca^{2+} and Cl^- were slightly higher at C site than R and I sites. The concentration of F^- was almost similar at all sites. The levels of other ions Ca^{2+} and Na^+ were lower at all the sites (Fig. 4). Among the three sites, average concentration of K^+, a biomass tracer (Lee *et al.*, 2010) was found to be slightly higher at I site than C and R site which may be due to the cooking activity that was going on in a marriage hall during the study period.

NO_3^- and SO_4^{2-} ions are generally found to be produced as secondary inorganic aerosol (SIA) during combustion and vehicular emissions (Seinfeld and Pandis, 2006). Good correlations were observed between SO_4^{2-} and NO_3^- (0.78, 0.87, 0.81 for R, C and I sites) indicating that they are formed via photochemical oxidation of SO_2 and NO_2 emitted from the common sources such as mobile, biomass burning and industries. The mass ratio of NO_3^-/SO_4^{2-} was used for identification of mobile and stationary sources of sulfur and nitrogen in the atmosphere (Arimoto *et al.*, 1996; Yao *et al.*, 2002). In the present study, NO_3^-/SO_4^{2-} ratio was ranged from 0.59 to 1.22 (mean 0.86 ± 0.22) at R site, from 0.69 to 1.36 (mean 0.92 ± 0.204) at C site and at I site, it was ranged from 0.48 to 0.89 (mean 0.70 ± 0.13). Mean ratio of NO_3^-/SO_4^{2-} was higher at C site followed by R site and then by I site according to the vehicular activities at the receptor sites. These mean ratios were higher than the data reported by Wang *et al.*, 2006 (0.64) indicating high influence of vehicular activities and low contribution from regionally transported sulphate from the stationary sources to the receptor sites. The other probable reason for higher sulfate and nitrate levels during the study period may be due to emission of SO_2 and NO_x from biomass burning which was prevalent at all the receptor sites (Gadi *et al.*, 2003).

Concentration of NH_4^+ was almost similar at R and I sites but lower at C site. Estimated high ammonium concentration at all sites could be due to several sources such as open sewerage system, biomass burning etc. (Hoek *et al.*, 2002).

Levels of Carbonaceous Matter

Fig. 5 shows the variations of OC and EC concentration levels in $PM_{2.5}$ at R, C and I sites. At R site, the average OC and EC concentrations were 23.5 ± 12.6 µg/m³ (8.9–43.2 µg/m³) and 8.5 ± 3.91 µg/m³ (3.4–15.2 µg/m³) respectively. At C site, average OC and EC concentrations were 24.2 ± 13.7 µg/m³ (7.3–41.8 µg/m³) and 10.1 ± 5.6 µg/m³ (3.5–16.7 µg/m³) while at I site, average OC and EC level were 21.9 ± 5.7 µg/m³ (13.5–28.4 µg/m³) and 8.5 ± 2.9 µg/m³ (5.6–12.3 µg/m³) respectively. The concentration of OC and EC levels followed the trend C > R > I. As compared to levels
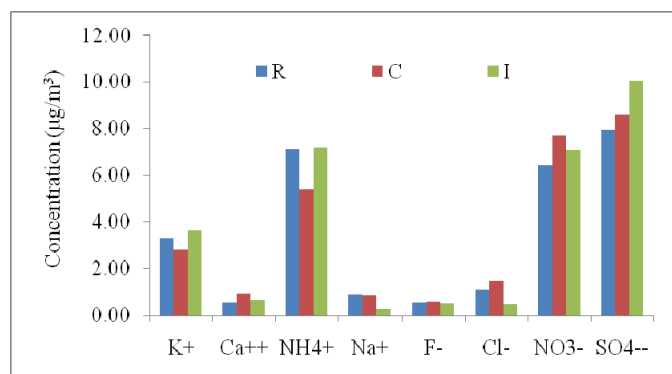
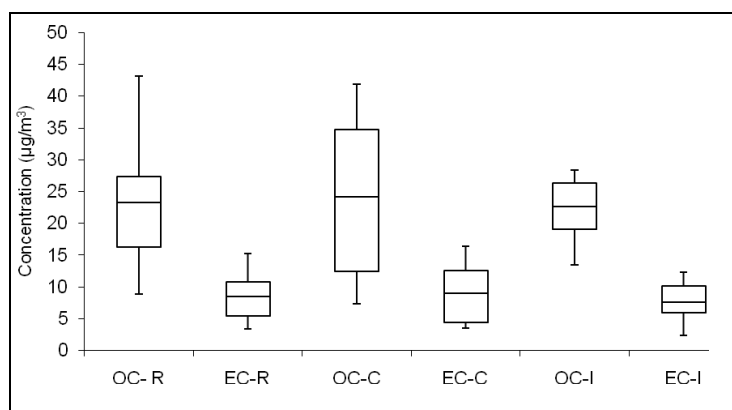**Fig. 4.** Average mass concentration of anions and cations in $PM_{2.5}$ ($\mu g/m^3$).

Fig. 5. Average mass concentration of OC and EC in $PM_{2.5}$ ($\mu g/m^3$).

at R and C sites, levels of OC and EC at I site were lower that may be due to the sampling height which was higher than the other two sites and secondly, no crowded traffic was observed in this locality due to the wider roads.

The relationship between OC and EC could help in identifying the origins of carbonaceous $PM_{2.5}$ (Turpin and Huntzicker, 1991; Chow *et al.*, 1996). Therefore, the mass ratio of OC to EC is used for studying emission and transformation characteristics of carbonaceous aerosol. In our study, correlations (r^2) of EC and OC were 0.82 (Y= 0.304 X + 1.429), 0.94 (Y= 0.394 X + 0.605), and 0.59 (Y= 0.391 X + 0.075) for R, C and I sites respectively. The high correlation in OC and EC in $PM_{2.5}$ showed that these are mainly attributed from common sources, such as motor vehicular emission and biomass burning.

Particles emitted due to biomass burning, consists of two large components i.e., OC and EC constituting nearly 50–70% mass of fine particles but the major contribution is of OC (Liousse *et al.*, 1996) while approximately 10% is composed of inorganic elements such as K^+, Ca^{2+} and SO_4^{2-} and NO_3^- and Cl^- etc. Therefore, K^+/OC and K^+/EC ratios can be used to characterize the emission sources (Andreae *et al.*, 1998).

The K^+/OC ratios at R, C and I sites determined in this study were 0.20, 0.20 and 0.17 respectively. K^+/OC ratios observed in previous studies were in the range of 0.08 to 0.10 for savanna burning, 0.04 to 0.13 for agricultural waste burning and 0.14 for biomass burning (Echalar *et al.*, 1995;

Andreae *et al.*, 2001, Zhu *et al.*, 2012). The ratios observed in our study were higher than these values. Similarly, K^+/EC ratios for R, C and I sites were 0.54, 0.47 and 0.45 respectively. The present results are within the range of previous studies, which reported the ratios from 0.20 to 0.69 for biomass burning aerosols (Andreae and Merlet, 2001; Ram *et al.*, 2010).

The ratios for SO_4^{2-}/EC and SO_4^{2-}/OC were found to be 1.08 ± 0.44, 1.34 ± 0.62 and 1.19 ± 0.38 and 0.45 ± 0.19, 0.60 ± .36, 0.48 ± 0.18 for R, C and I sites respectively. Similarly, NO_3^-/EC and NO_3^-/OC ratios were found to be 0.40 ± 0.11, 0.55 ± 0.21, 0.34 ± 0.22 and 0.93 ± 0.23, 1.23 ± 0.32, 1.23 ± 0.43 for R, C and I sites respectively. These ratios of NO_3^-/EC and NO_3^-/OC are higher than those calculated from road tunnel aerosols (0.1 for NO_3^-/EC and 0.09 for NO_3^-/OC) (Gillies *et al.*, 2001) indicating that vehicular as well as biomass burning activities are contributing at all receptor sites.

Chemical Mass Closure

Chemical mass closure method is used for the determination of relative contributions of quantified chemical components and their sources to the fine particulate mass. The gravimetrically-measured particulate mass of $PM_{2.5}$ is reconstructed from the sum of quantified chemical components. In this study, mass balance for the averages of OC, EC, NH_4^+, NO_3^-, SO_4^{2-}, trace metals and unexplained mass was reconstructed for $PM_{2.5}$ for three sites and are

depicted in Figs. 6(a)–6(c). The result showed that concentrations of OC, EC and SO_4^{2-} were higher at all the three sites. Contributions of OC to $PM_{2.5}$ were 26.7, 27.6 and 26.5% whereas EC were 11.1, 9.6 and 8.9% at R, C and I sites respectively. The SIA i.e., SO_4^{2-}, NO_3^- and NH_4^+ together contributed 28.2, 26.2 and 29.0% to $PM_{2.5}$ at R, C and I sites respectively. The other ions such as K^+ shared nearly 4% while, Ca^{2+} ions shared about 1% of $PM_{2.5}$ mass at all the sites. The trace metals contributed 3.5, 3.2 and 2.6% at R, C and I sites respectively. The average unexplained portion of $PM_{2.5}$ mass at R, C and I sites were 22.2, 25.6 and 26.5% respectively.

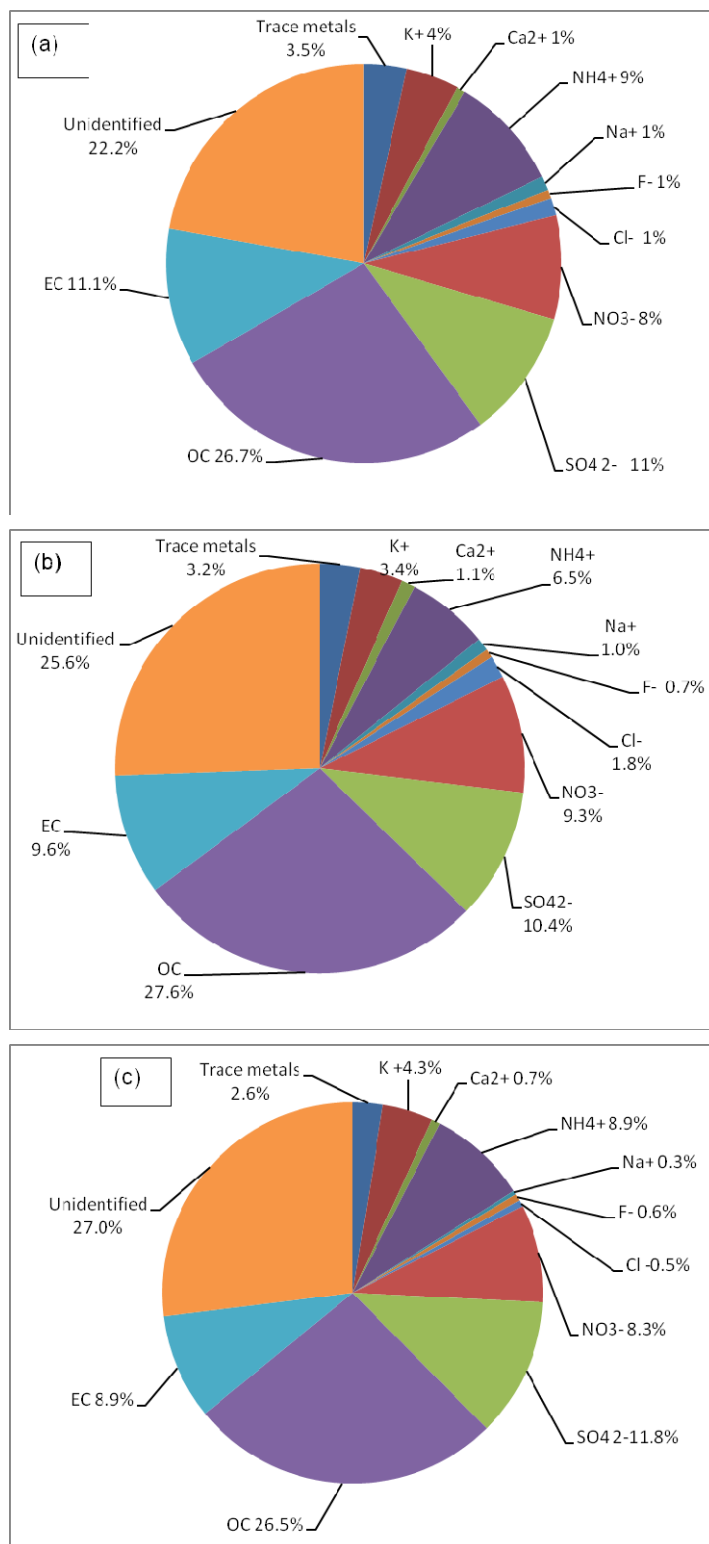**Fig. 6.** Mass closure of $PM_{2.5}$ at (a) R, (b) C and (c) I sites.

As per the findings of Rastogi and Sarin (2009) and Kumar and Sarin (2009) in western India, water-soluble ionic composition constituted 50% of $PM_{2.5}$ mass during winter, with dominant contribution from SO_4^{2-} and NH_4^+ for which biomass burning and fossil-fuel emissions were major sources. Whereas a study carried out by Rengarajan *et al.*, (2011) over an urban site in a semi-arid region of western India during winter, found nearly 29% water soluble inorganic constituents indicating formation of SIA. In this study, water soluble inorganic components were 35-37% out of which NH_4^+, SO_4^{2-} and NO_3^- were dominant species due to traffic and other local anthropogenic activities such as biomass burning.

Source Apportionment Results of $PM_{2.5}$

Source profiles of gasoline/diesel vehicles, biomass burning, paved and unpaved road dust (re-suspended dust), secondary sulphate, secondary nitrate, DG set, coal and industrial combustion were included in CMB model. These source profiles were selected on the basis of primary survey around receptor sites of 2×2 km grids. The performance of CMB results were evaluated by the performance indicators as percentage mass explained, R^2 and $\chi 2$ values while quantification of major contributing sources to $PM_{2.5}$ mass and estimated percent contributions are presented in Fig. 7. The R^2 and $\chi 2$ values for sites R, C and I were 0.86, 0.87 and 0.91 and 2.43, 2.65 and 2.13 respectively. The mass percentages calculated were found to be 88, 85 and 87% for the sites R, C and I respectively. Statistical parameters of the model applied to $PM_{2.5}$ for three sites were within the acceptable intervals indicating a good performance of the model application.

Contribution of mobile source at R site was nearly (57.0%) followed by secondary aerosol (16%), open burning (15.1%), resuspended dust (6.0%) and then unidentified remain as 6.0% to $PM_{2.5}$. At C site, contribution of mobile source was 62%, secondary aerosol was 12.0%, open burning was 11.0%, resuspended dust as 10.0% and unidentified remain as 5.0%. At I site, contribution of vehicular

emission was 65% followed by secondary aerosol (16.0%), resuspended dust (9.0%), open burning (7%), and finally unidentified remain as 3.0%. Findings showed that sources at all the three sites were most consistent but percent contribution of these sources varied among the sites as per the intensity of ongoing activities at receptor sites.

CONCLUSIONS

$PM_{2.5}$ concentrations in Nagpur city were found to be 67.1 ± 22.1, 96.5 ± 21.6 and 85.3 ± 16.2 μg/m³ at sites R, C and I respectively. Levels at all the sites were exceeding the daily NAAQS promulgated by CPCB. The most abundant chemical species were OC, EC, SO_4^{2-}, NO_3^-, NH_4^+, K^+ and trace metals (Al, Fe, Si, Mg, Cu) at all the sites. Source profiles developed in India for non-vehicular and vehicular sources used in CMB along with ammonium sulfate, and ammonium nitrate profiles based on the calculated mass balance for the source apportionment of $PM_{2.5}$ has resolved four sources major being the mobile sources, followed by SIA, biomass burning and finally by resuspended dust. Sources were consistent at all the three sites but percent contribution of these sources varied among the sites as per the intensity of ongoing activities. No doubt, this study would serve as a baseline observation to prepare action plans for controlling the identified sources.
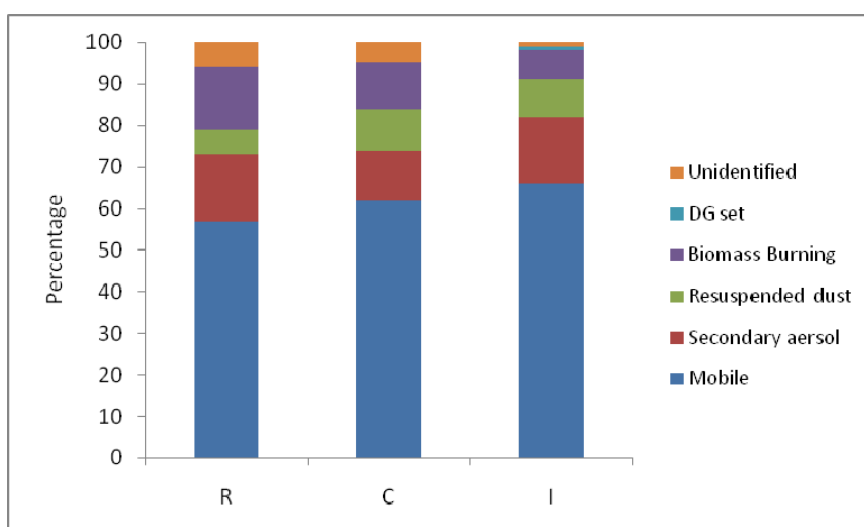
ACKNOWLEDGMENTS

Fig. 7. Source apportionment results of $PM_{2.5}$.

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

Supplementary Table: Source Profiles (percentage) developed by CPCB and ARAI used in this study.

Parameter/ Sources	Open Burning	Unpaved Road Dust	Paved Road Dust	Dieseal Geneartor	Soil	Gasoline vehicles	Diesel vehicles	Ammonium Sulphate	Ammonium Nitrate
Al	0.004	0.122	0.136	0.337	0.125	0.03	0	0	0
Uncecert	0	0.009	0.01	0.025	0.009	0.16	0.02	0	0
Ba	0.016	0.02	0.032	0.041	0.03	1.53	0.58	0	0
Uncecert	0.001	0.002	0.003	0	0.002	0.07	0.01	0	0
Cd	0.001	0.002	0.008	0.029	0.001	0.22	0.01	0	0
Uncecert	0	0	0.001	0.004	0.001	0.05	0	0	0
Cr	0.003	0.008	0.126	0.387	0.01	0	0	0	0
Uncecert	0	0.001	0.01	0.03	0.002	0.01	0	0	0
Cu	0	0.017	0.028	0.449	0.013	0.02	0.01	0	0
Uncecert	0	0.002	0.003	0.034	0.002	0.01	0	0	0
Fe	0.486	6.715	6.523	57.624	6.343	0.15	0.02	0	0
Uncecert	0.035	0.476	0.463	4.079	0.45	0.02	0.02	0	0
Mg	0.001	1.791	1.318	0.175	1.285	0.02	0	0	0
Uncecert	0.001	0.127	0.094	0.017	0.092	2.42	0.24	0	0
Mn	0.003	0.116	0.128	0.001	0.164	0.067	0.002	0	0
Uncecert	0	0.008	0.009	0.337	0.012	0.017	0.002	0	0
Mo	0.001	0.003	0.01	0.056	0.033	0.006	0.001	0	0
Uncecert	0.001	0.001	0	1.114	0.006	0.028	0.003	0	0
Ni	0.001	0.021	0.067	0.264	0.037	0.086	0.003	0	0
Uncecert	0.005	0.009	0.021	6.572	0.029	0.007	0.001	0	0
Pb	0.017	0.01	0.184	0.07	0.051	0.11	0.01	0	0
Uncecert	0.006	0.008	0.029	0.5	0.021	0.01	0	0	0
Si	0.061	0.032	0.099	0.26	0.104	0	0	0	0
Uncecert	0.005	0.004	0.012	3.197	0.015	0	0	0	0
Zn	0.083	0.009	0.094	0.82	0.037	0.68	0.25	0	0
Uncecert	0.006	0.001	0.007	0.57	0.003	0.01	0	0	0
F^-	0.048	0.06	0.645	0.02	0.09	0.35	0.06	0	0
Uncecert	0.005	0.006	0.05	0.11	0.02	0.41	0.14	0	0
Cl^-	0.262	0.087	0.427	1.67	0.232	3.63	0.34	0	0
Uncecert	0.05	0.063	0.151	3.43	0.167	0.79	0.12	0	0
NO_3^-	0.014	0.019	0.042	0.24	0.051	0.85	0.18	0	77.5
Uncecert	0.009	0.014	0.03	0.69	0.037	0.22	0.02	0	7.5
SO_4^{2-}	0.251	0.259	1.976	0.18	0.149	4.54	0.83	72.7	0
Uncecert	0.022	0.025	0.153	0.45	0.03	0.71	0.08	7.27	0
K^+	27.06	0.157	0.346	0.9	0.419	1.2	0.54	0	0
Uncecert	2.02	0.113	0.249	4.99	0.301	1.77	0.09	0	0
Ca^{2+}	1.754	2.87	3.239	0.53	1.935	2.83	0.87	0	0
Uncecert	0.227	0.37	0.622	8.84	0.633	0.85	0.12	0	0
NH_4^+	0.105	0.088	0.193	1.1	0.233	0.27	0.02	27.3	22.5
Uncecert	0.041	0.063	0.139	3.13	0.167	0.15	0	2.73	2.25
Na^{2+}	0.107	0.091	0.455	1.56	0.55	2.03	0.42	0	0
Uncecert	0.09	0.143	0.327	6.75	0.395	3.48	0.16	0	0
OC	52.059	3.643	8.419	6.594	4.124	52.05	49.27	0	0
Uncecert	2.252	0.182	0.421	3.099	0.206	4.31	4.01	0	0
EC	1.44	1.432	2.551	12.934	1.528	6.58	21.57	0	0
Uncecert	0.062	0.072	0.128	5.73	0.076	0.56	1.79	0	0

Supplementary Table: CMB performance results for R, C and I sites in terms of standard error, T_{stast}.

Source	R			C			I		
	SCE** ($\mu g/m^3$)	Std Error	T stast	SCE ($\mu g/m^3$)	Std Error	T stast	SCE ($\mu g/m^3$)	Std Error	T stast
Biomass burning	9.90	0.83	11.94	7.27	0.93	7.79	7.10	1.80	3.80
paved road	4.72	0.45	10.40	5.60	0.78	7.09	8.82	1.60	5.30
Diesel vehicle	22.78	2.59	8.78	36.08	2.45	14.72	42.37	6.80	6.20
DG set*	-	-	-	-	-	-	0.77	0.17	4.60
Gasoline Vehicle	13.35	2.60	5.13	24.25	2.12	11.43	21.89	3.02	7.20
Ammonium sulphate	5.49	1.00	5.49	9.95	1.34	7.42	8.31	1.37	6.00
Amm nitrate	4.96	0.46	10.90	1.65	1.56	1.05	7.73	0.72	10.70

* Diesel generator
** Source contribution estimate