Source Identification and Distribution of Toxic Trace Metals in Respirable Dust (PM 10) in Brasscity of India

Anamika Tripathi[1] and Atul Kumar[2]

[1] Hindu College

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Abstract

This study assessed the concentration of PM10 and trace metals at six sites with different land uses during the period of one year. Metals concentrations of PM10 were analyzed using ICP-OES. Highest concentrations of PM10were recorded in winter and lower in monsoon at all the study sites. The concentrations of trace metals in PM10 were observed in the following order: Zn > Fe > Cu >Al > Pb > Cr > Mn > Cd > Ni. Overall concentration of PM10 and heavy metals was found highest at industrial sites than the vehicular, commercial and residential sites shows the greater contribution of industrial and combustion process. Univariate (correlation study) and Multivariate statistical analysis were adopted including; factor analysis and enrichment factor analysis to identify the sources and their contribution to PM10. The major source of airborne trace metals identified were brassware industries, illegal e-waste burning automobile emissions and combustion processes.

Index terms— respirable dust, trace metals, principal component analysis, enrichment factor analysis, industrial activities, e-waste burning, automobile emissions.

1 Introduction

tmospheric particular matter is considered as a prime pollutant of concern for urban cities, not only because of the adverse health effects, but also for the reducing atmospheric visibility (Grieken & Delalieux, 2004;Quinn et al., 2005). On a Global scale, particular matter (PM) also influences directly and/or indirectly the Earth's radiation energy balance, and can subsequently impact on global climate change **??**IPCC, 2001). Atmospheric particulates are reported to affect ecosystems (Niyogi et al., 2004) and materials adversely. A number of studies have been undertaken focusing on the characteristics of aerosols in megacities of the world including Beijing , Colombo, Oxford, Amsterdam, Athena, Jeddah etc. (Sun et al., 2004; **??**eneviratne et al., 2011;Wojas and Almquist, 2007;Vallius 2005;Chaloulakou et al., 2003;Khodeir et al., 2012).

PM 10 particles (the fraction of particulates in air of very small size (<10?m) are of major current concern, as they are small enough to penetrate deep into the lungs and so potentially pose significant health risks (Begum et al., 2004;Artinano et al., 2007;Guttikunda et al., 2014). The results of the long -term studies confirm that the adverse health effects are mainly due to microns in diameter, PM 10 (Schwartz et al., 1996). The particulate may include a broad range of chemical species, ranging from metals to organic and inorganic compounds (Tsai and Cheng, 2004;Park and Kim, 2005). Among the inorganic compounds, most important ones are the trace metals, which are emitted by various natural and anthropogenic sources such as crustal materials, road dust, construction activities, motor vehicles, coal and oil combustion, incineration and industrial metallurgical process (Quiterio et al., 2004;Shah et al., 2006;Park et al., 2008;Shah and Shaheen, 2010;Cheng et al., 2011). Industrial metallurgical process is regarded as one of the most important anthropogenic trace metal emission sources (Zheng et al., 2010) and produce the largest emissions of trace metals as As, Mn, Co, Cd, Cu, Ni and Zn **??**Vassilakos et al., 2006; Van et al., 2014).

Airborne particulate matter with elevated metals may have a serious impact on human health which mostly includes respiratory disease, lung cancer, heart disease and damage to other organs (Magas et al., 2007;Liu et al., 2009;Mavroidis and Chaloulakou, 2010). Within the European programme for monitoring and evaluation of the long -range transmission of air pollutants (EMEP), measurements of PM 10 and heavy metals, are highly toxic species have been introduced. These observations are influencing the environmental legislative authorities all over the world to update and modify their air quality standards ??WHO, 2006; ??uropean Commission, 2004; ??SEPA, 2008). The recommended guidelines for maximum PM 10 concentrations are 50µgm -3 (24-h average) where as 20 µgm -3 for the annual average concentration.

Moradabad, the 'Brass city of India' is the second most populated city of state Uttar Pradesh and the most significant commercial centre of Northern India. More than 80 % of the total production of brass souvenirs and utensils of India is from Moradabad region alone. It is one of the largest producing and exporting center of brass-wares in India. The growth of the city over the last thirty years has been rapid and (Pathak et al., 2008) and illegal e-waste burning units in dense residential areas (Figure ??) while mobile source of pollution includes all forms of transportation. Vehicle fuels used in Moradabad are mainly unleaded gasoline and diesel although some lead is still permissible. Pollution assessment in this area is important since air quality has a major influence on workers of the industries and inhabitants living around the area.

The objective of the study was to assess air quality and to identify the main source by multivariate receptor modeling (PCA), enrichment factor (EF) calculation and analysis of meteorological effects. Anthropogenic enrichment of trace metals in atmospheric particulates were also envisaged along with the comparative evaluation of the estimated metal levels with those reported from other areas around the world. The results could be used as the baseline data for analysis of health risk due to inhalation of respirable dust (PM 10), and to provide scientific evidence for setting up an air pollution control.

2 II.

3 Material and Methods

4 c) Analytical Technique

Before and after the sampling procedure, filters were kept for 48 h in desiccators in an environmentally conditioned room with a RH of 45±5% and a temperature of 20±2 o C before being weighed by a microbalance (Sartorius BP160P). The difference in initial and final weight (gravimetric analysis) of the filter paper gave the total quantity of PM 10 collected over the 24 hours period. The values of PM 10 were reported in µgm -3 . For analysis of metallic elements, total 72 square of 1×1 ins diameter (6 locations + 1 control/blank) of the fiber filter paper covered by particulates digested with nitric acid and perchloric acid in a ratio 1:3 on a 140 0 C hot plate till white fumes arose. Residues were then redissolved by 0.1M hydrochloric acid and the content was filtered through Whatman Filter no. 42 and finally made-up to 25 mL by double distilled water. The filtrate of each sample was examined for the concentrations of heavy metals by using Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES; Spectro Analytical Instruments, West Midlands, UK) collected for each site at Metal Handicraft Service Centre, Peetal Nagari, Moradabad (Ministry of Textiles, Govt. of India). To get the final concentration results of the blank samples are subtracted from the exposed samples. For each of the metals the concentration of metals in the samples is then multiplied by the sample volume (i.e. 25 mL) to get the mass of each metal. These values are subsequently divided by corresponding total volume of sampled air to get the concentration of metals in the sampled air.

5 d) Data Analysis Techniques

Obtained data were processed for statistical analysis including univariate and multivariate methods. Basic statistical parameters such as mean and standard deviation are computed along with correlation analysis, while multivariate statistics in terms of Principal Component Analysis (PCA) as given by Lee and Hieu, 2011 . Calculation of Enrichment Factors (EFs) was also performed using the SPSS version 16.0 statistical software. a flow rate of 1.0-1.5 m 3 /min and the monitoring of III.

6 Results and Discussion

7 a) PM 10 concentrations

The site-to-site seasonal comparison of PM 10 mass concentration is statistically presented in Table ??. The range of mass concentrations varied considerably over time from 71 to 181 µgm -3 at Buddhi Vihar (SI, Residential) from 45 to 238 µgm -3 at Buddh Bazar (SII, Commercial), from 61 to 213 µgm -3 at Kapoor Company (SIII, Traffic density), from 54 to 174 µgm -3 at PTC (SIV, Residential), from 109 To 244 µgm -3 at Peetal Nagari (SV, Industrial) from 99 to 213 µgm -3 at Mughalpura (SVI, Industrial and illegal e-waste burning) and all the monthly mean values are found more than the recommended concentration of NAAQS (60 µgm -3) except SII (45 µgm -3) and SIV (54 µgm -3) in the month of July and August respectively but all the values are high than the recommended concentration of WHO for PM 10 (20 µgm -3) at all the study area (WHO, 2006). The annual average concentrations at each location were 124±43, 137±59 and 131±49, 118±45, 193±43, 182±39 µgm -3 respectively. The lowest concentration was found at PTC (SIV) the residential area which surrounds by greenery and the highest concentration was found at industrial area of Peetal Nagari characterized by industrial activity and soil dust.

The annual average values obtained from Peetal Nagari (SV) and Mughalpura (SVI) were higher than the USEPA recommended annual PM 10 ambient air quality standard, i.e. 150 µgm -3 (USEPA, 1999). However the value at the Buddhi Vihar (SI), Buddh Bazar (SII), Kapoor Company (SIII) and PTC (SIII) is lower than the USEPA PM 10 standard. The higher PM 10 concentrations at site Peetal Nagari and Mughalpura may reflect a significant contribution of anthropogenic sources compared to the other sites. High ambient PM 10 mass concentration peaks occurred at industrial sites, suggesting that contribution of stationary industrial emission were more important than the contribution of mobile sources even in areas with heavy traffic (Chen et al., 2008;Roy et al., 2012).

A seasonal variation was found as higher concentrations for PM 10 occurred in winter period (November-February) at all the study area than those of summer (March-June) which could possibly be attributed to the higher traffic density and combustion of fossil fuel for heating during winters as well as prevailed meteorological conditions (Table **??**). The winter months of are relatively calm than other months thereby causing slow dispersion of pollutants generated and helps in buildup of pollutants in vicinity of the pollutant sources. Lower average mixing height in winter season results in less volume of troposphere available for mixing and hence higher PM 10 concentrations. The low temperature during winters lead to more energy consumption for industrial purpose resulting in emitting more emission of primary PM from the industrial sources (Lee & Hieu, 2011). Almost at all the sampling sites the concentration of PM 10 was found lowest (Table **??**) in monsoon season (July-October) which usually has large amounts of precipitation and high relative humidity (Table **??**). These meteorological conditions such as increased rainfall and humidity during the monsoon period can greatly decrease PM concentrations in ambient air via rainout or washout mechanism (Pillai et al., 2002;Glavas et al., 2008).

Summer months (March-June) shows comparatively lower values than the winter may be due to high wind speed, causing dispersal of pollutants. Thus the difference in PM concentrations between the three seasons can be explained by the difference in weather pattern or meteorological conditions for each specific season.

Moradabad's PM levels were compared with those in different urban locations across Europe and Asia (Table 3). The average concentration of PM 10 i.e. 148 µgm -3 was recorded during this study which was significantly higher than the other studies of the world. It is near about the concentration found in Hyderabad i.e.135 µgm -3 (Sun et al., 2004). As the maximum average mass concentration during winter for PM 10 were 446, 573, and 631 µgm -3 at the traffic, industrial and residential sites in Beijing, respectively. These value were compared to the maximum PM 10 concentration of our sites which were 179, 203 µgm -3 at SI and SIV (compared to the residential site) respectively, 203, 213 µgm -3 at SII and SIII (compared to the traffic site) and 234, 213 µgm -3 at SV and SVI (compared to the industrial site). All these concentrations were less from the findings of Sun et al., 2004. As Moradabad is a small city comparatively to highly developed megacity Beijing, hence the sources are more in Beijing in comparison to Moradabad.

8 b) Trace Metal Concentrations

Considerable differences were noted with respect to metal content in samples of PM 10 from Buddhi Vihar (SI, Residential), Buddh Bazar (SII, Commercial) Kapoor company (SIII, Traffic) PTC (SIV, Residential), Peetal Nagari (SV, Industrial) and Mughalpura (SVI, Industrial and illegal e-waste burning). Heavy metals such as Fe, Al, Cu, Zn, Mn, Ni, Pb, Cr, and Cd concentration along with standard deviation were displayed in figure 2 at all the sampling sites. Among the trace metals Zn contributed the maximum concentration with annual average of 11.84 µgm -3 followed by Fe (9.41 µgm -3), Cu (7.57 µgm -3), Al (5.74 µgm -3), Pb (1.99 µgm -3), Cr (0.21 µgm -3) Mn (0.11 µgm -3), Cd (0.09 µgm -3) and Ni (0.01 µgm -3). Among all the six monitoring sites the highest concentration of Fe (18.43 µgm -3), Al (10.08 µgm -3) Cu (15.23 µgm -3) and Cr (0.41 µgm -3) was observed at Mughalpura (SVI) followed by Peetal Nagari (SV) 17.07, 9.88, 14.84, 0.39 µgm -3 respectively while Zn (21.09, 21.21 µgm -3) and Ni (0.031, 0.034 µgm -3) was found almost same at both the site. The Mughalpura and Peetal Nagari sites were surrounded by many small and large scale brassware industries. In these industries, Brass (60% Cu and 40% Zn) and German silver (55% Cu, 35% Zn and 10% Ni) are the main alloys used for moulding purpose in making brassware items and other utensils in Moradabad. Brassware industries which are specialized in cutting, grinding, scraping, polishing etc. are the major cause of high concentration of these metals **??**Tripathi et Peetal Nagari (industrial site), situated along the major road connected to Delhi is a major exporting centre of brasswares so the vehicular traffic as well as industrial activity could be the major source of Cu, Zn and Cr. As Cu is associated mainly with industrial activities, road traffic (diesel engine and wearing of brakes) could be the most important source in urban area. Zn is reliable tracer of unleaded fuel and diesel oil powered motor vehicles emissions (Monaci et al., 2000) and besides, it could be released in large amounts from tired friction or various industrial activities. Use of oil lubricants at the service centers, tire abrasions and vehicle exhausts are the possible sources of Cr at the study areas. Presence of such sources and their association with increased Cr and Zn concentrations comply with the findings of Karar et al., 2006 andBhaskar et al., 2008. Highest value of Pb (2.72 µgm -3) and Cd (0.21 µgm -3) was observed at Buddh Bazar (SII), a very busy commercial site along with vehicular activity throughout the day and night followed by Kapoor Company (SIII), 2.5, 0.17 µgm -3 respectively. The concentration of Pb in higher amount is mainly due to traffic volume (Tripathi, 1994;Xia and Gao, 2011). As lead pollution due to leaded gasoline still occurs in few cities (Prajapati et al., 2009;Andra et al., 2011). The major source of human lead accumulation in developing countries was found to be airborne lead and 90 percent of which comes from leaded gasoline ??MECA, 2003). Cadmium, one of the most dangerous pollutants for organism, is mainly derived from combustion of accumulators and carburetors of vehicles (Divrikli et al., 2006). It is a major industrial pollutant particularly in areas associated with smelting of zinc and heavy road traffic (Hassan et al., 2009). Mn which is mainly derived from the anthropogenic activities found highest mean value at Peetal Nagari followed by Kapoor Company. The residential sites showed comparatively the lower concentration.

9 c) Correlation Analysis

Correlation coefficient was used to establish interrelationship between metals (Table 4). The strong correlation (0.754, 0.729) was found between Fe-Al and Cu-Zn respectively in the study area. The significant correlation was found between Fe with Cu (r=0.679), Zn (r=0.695), Ni (r=0.625) and Cr (r=0.504). It is also found significant for Al with Cu (r=0.688), Zn (r=0.581), Ni (r=0.643) and Zn-Ni (r = 0.541), Mn-Ni (r=0.60). It may be due to the industrial and anthropogenic activities like burning of fossil fuel. Zn-Cr (r=0.433), Al-Cr (r=0.419), Cu-Ni (r=0.490), Pb-Cd (r=0.421), Cr-Cd (r=0.41) showed the moderate correlation while the negative correlation was found between Ni-Pb and Ni-Cd. Based on the correlation study, it may be concluded that Fe, Al, Cu, Zn, Ni and Cr were contributed by some common sources, probably by industrial and anthropogenic Sources.

10 d) Factor Analysis

The principal application of factor analysis is to reduce the number of variables. This method focuses on cleaning up the factors. PCA was applied to determine the correlation between pollutants and to identify the source profile of heavy metals in PM 10 . Table 5 describes the Principal Component (PC) loadings for the metal data of the study period with corresponding eigen values and variances. Based on this matrix three new sets of synthetic variables (Principal Component) were obtained. For interpreting of the data the method of Kaiser Criterion (Kaiser, 1960) is followed which retain only those factor having eigen value greater than 1 has been used for further interpretation. Factor loading > 0.71 are typically regarded as excellent and < 0.32 as very poor (Nowak, 1998). The Ist PC explains 40.113% of data variance and it is characterized by Fe, Al, Cu, Zn and Ni. These heavy metals are mainly related to the Industrial emissions, especially the metallurgical/ electroplating and e-waste burning units located in the industrial area of the city (Wang et al., 2001;Quiterio et al., 2004;Shah and Shaheen 2008). The IInd factor characterized by Pb and Cd. These heavy metals are well known to be associated with the automobiles ??Ayras and Kaushilina, 2000). The IIIrd component is characterized by Mn, mainly derived from anthropogenic activities. The extracted components explain nearly 74.31% of the variability in the original 9 variables. The number of eigen values can be estimated from a scree plot demonstrated in figure ??. As shown in this figure, the eigen value sharply decrease within the first three components and then slowly stabilize for the remaining ones.

11 e) Enrichment Factor Analysis

Enrichment factor (EF) analysis was used to differentiate between the metals originating from human activities and those of natural origin and to assess the degree of anthropogenic influence. By convention, the average metalal concentration of the natural crust is used instead of the continental crust composition of the specific area, as detailed data for different areas are not easily available. There is no rule for the reference metal choice and Si, Al, and Fe have been used as the most common metals for this purpose (Lee and Hieu 2011). In this study, Fe used as the reference metal with upper continental crustal composition given by Taylor and McLennan (1985). Since iron (Fe) has been used as a reference metal for an EF evaluation, assuming that the contribution of its anthropogenic sources to the atmosphere is negligible (Nazir et al., 2011). The enrichment factor is calculated through the following equation:(E/R) air sample EF = (E/R) crust

EF represents the ratio of the fraction of the metal E with respect to reference metal R in the samples. (E/R) sample to the fraction of E with respect to the same R in the crust (E/R)crust. The EFs of individual metals are shown in Figure ??. According to the degree of enrichment, the metals were grouped as follows:

? Highly enriched (EF > 100) included Pb, Zn, Cu and Cd. ? Moderately enriched (EF between 10 and 100) none of them. ? Less enriched (EF less than 10) included Al, Mn, Ni and Cr.

In the present study, large variation of EF values was found for different metals in the respirable dust. Amongst these EFs of Cd and Cu are the highest followed by Zn and Pb. The higher EF values of these metals showed the anthropogenic sources (industrial, automobile and combustion emission) contributed a substantial amount of the metals in atmospheric particulates, which otherwise were difficult to justify on the basis of normal crustal weathering process. In contrast the less enriched metals were dominantly derived from earth crust, and re-suspension of soil dust. On the whole, all metals revealed EF greater than unity, thus predominantly contributed by the anthropogenic source.

IV.

12 Conclusion

The study area covers a substantial portion of Moradabad city. Overall site specific analysis of PM 10 data reveals that Peetal Nagari is the most polluted area in terms of dust loading with a maximum concentration of 234 µgm -3 followed by Mughalpura. The concentrations of PM 10 in winter was higher than those in summer and monsoon. Increased energy uses, low temperature and low mixing height contributed to increasing PM concentrations in winter months while increased rainfall precipitation in monsoon period greatly contribute to decrease PM level. The concentration of PM 10 in Moradabad recorded high than those from the other sites in Europe and Asia except Beijing. The characterization of trace metal sources in the study area is quite challenging due to a large number of industrial and urban sources. High concentration of Zn, Cu, Cr and Ni at industrial site (Peetal Nagari and Mughalpura) was found mainly due to its use in brasswares and electroplating. Pb and Cd was found highest at traffic and commercial sites (Kapoor Company and Buddh Bazar) was due the vehicular emission and combustion process but the high concentration in Mughalpura area is mainly due to burning of e-waste near this site, which is brought from Delhi in an illegal way. Focusing our attention on metal source characterization, the multivariate techniques allowed us to identify three source components. The PC I (40.113%) is characterized by Fe Al, Cu, Zn and Ni which represents industrial emission and combustion of fossil fuel. The PC II (20.470%) is associated with vehicular traffic emission and characterized by Pb and Cd. The PC III (13.736%) is identified as anthropogenic sources and characterized by Mn. Calculation of enrichment factors (EF) of the trace metals showed high enrichment of Cd, Cu, Zn and Pb, indicating heavy contamination by anthropogenic sources. These results are also supported by correlation study. Hence we conclude that in the investigated areas the level of some trace metals are very high and the level of PM 10 was also found higher than the NAAQS and WHO standard even in the residential areas. Due to high pollution level the people of the study area are suffering from many diseases related to air pollution. This suggests that future strategies for air quality control on a local scale have to take into account not only the amount of atmospheric particles, but their chemical composition as well.

V.

1234

Figure 1: Figure 1 :Figure 2 :Figure 3 :Figure 4 :

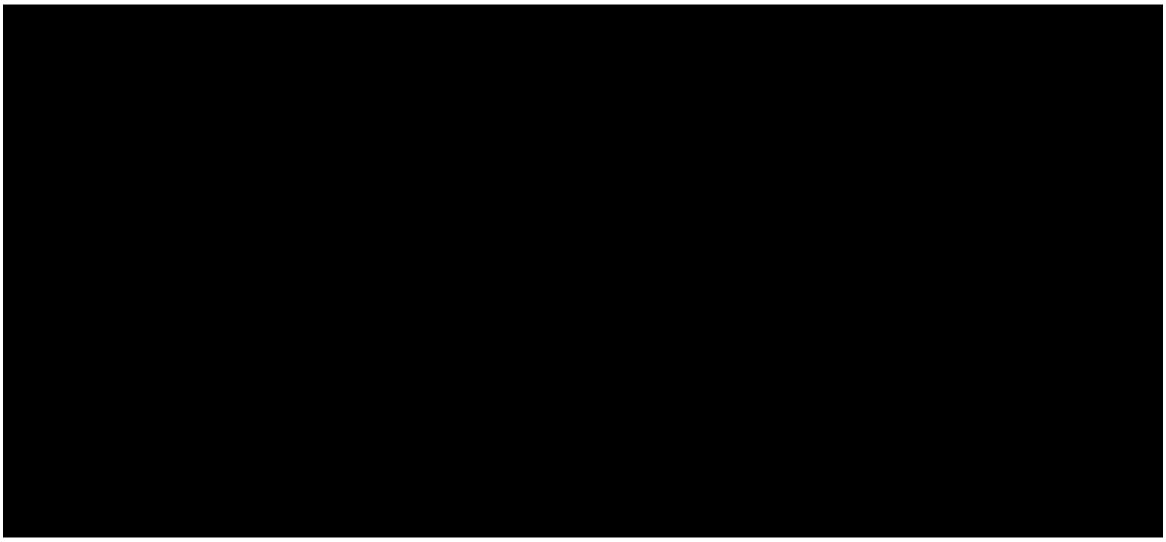

Figure 2:

Figure 3: A

Figure 4:

Figure 5:

3

Figure 6: Table 3 :

4

Figure 7: Table 4 :

5

Rotated Component Matrix a			
		Component	
Elements	PC I	PC II	PC III
Fe	0.856	0.150	0.239
Al	0.852	-0.044	0.281
Cu	0.835	0.147	0.016
Zn	0.777	0.471	0.012
Mn	0.078	0.017	0.935
Ni	0.824	-0.170	-0.019
Pb	0.072	0.783	-0.020
Cr	0.397	0.449	0.463
Cd	-0.033	0.854	
Eigen values	3.610	1.842	1.236
% Variance	40.113	20.470	13.736
%Cumulative variance	40.113	60.584	74.319
Possible Sources	Industrial emission	Vehicular emission	Anthropogenic activity

Extraction Method: Principal Component Analysis.
Rotation Method: Varimax with Kaiser Normalization.

[Note: r-values shown in bold are significant at p?0.001]

Figure 8: Table 5 :

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