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Appraisement of Heavy Metals in Respirable Dust (PM_{10}) Around E-Waste Burning and Industrial Sites of Moradabad: Accentuation on Spatial Distribution, Seasonal Variation and Potential Sources of Heavy Metal

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Abstract: *The primitive e-waste recycling techniques along with industrial activities are among the most destructive activities in Moradabad, threatening the health, polluting the air and water and harming the land. The variations in air quality in terms of respirable dust i.e. PM_{10} and their heavy metal content around e-waste burning and industrial area were evaluated over the period of April 2015- September 2015 at five sites during different seasons i.e. summer and monsoon. Metals concentrations of PM_{10} were analyzed using ICP-OES. Highest concentrations of PM_{10} were recorded in summer and lower in monsoon at all the study sites. Mean concentration of PM_{10} and heavy metals was found highest at industrial sites and e-waste burning site than residential sites. The mean concentrations of heavy metals in PM_{10} were found in the order of Cr < Ni < Cd < Pb < Cu < Zn. Multivariate statistical analyses were adopted including; principal component analysis (PCA) to identify the major sources of air pollutants in the area. The major sources contributing to air pollution in Moradabad were e-waste processing and industries related activities and secondarily vehicular emissions, while wind–blown dust through unpaved roads also contributed to some extent. The exceedance of PM_{10} concentration and their heavy metal content than the standards imposed negative effects to the environment, which made the local residents face with the serious heavy metal exposure.*

Keywords: *Reparable dust (PM_{10}), heavy metal, E-waste, Industrial activities, Human health.*

I. Introduction

Moradabad is the largest centre of producing and exporting of brassware's in India so it is known as the *"Brass city of India"* or *Peetal Nagri*. About some estimation, 80 % of the total production of brass memento, contraption and utensils of India is from Moradabad region alone (Pal *et al*., 2014). Although it is famous for brassware industries but now a day it has emerged as an important destination for illegal burning of e-waste generated in India as well as other countries. Electronic waste (e-waste) comprises waste electronics/electrical goods such as computers, printers, mobile phones, TVs etc. that are not fit for their originally intended use or have reached their end of life. It is much more hazardous than many other municipal wastes because it contains thousands of toxic ingredients includes heavy metals such as lead, mercury, cadmium, arsenic, chromium and harmful chemical such as polychlorinated biphenyl (PCB), brominated flame retardants (BFR) (Singh *el al*., 2015).

It is estimated that about half the printed circuit boards used in the appliances in India end up in Moradabad (Down to Earth, 2015). The circuit boards are sourced from e-waste such as computer monitors, television, cell phones and other electrical appliances. Recyclers in Moradabad buy PCB (printed circuit board) from Delhi, Kolkata, Chennai and other parts of India. Global recession and the decreasing demand for brass products in the West forced the manufacturers and the handicraft workers to adopt the hazardous e-waste recycling to earn their livelihood. The handicraft workers are experts in crafting metals but now are experienced at extracting metals such as copper, aluminium, gold and silver from electronic products (CSE, 2015; Singh *et al*., 2016). Currently, e-waste recycling in Moradabad is completely undertaken by the unorganized sector. The processes involved in e-waste recycling by the informal sector are highly polluting and are not only hazardous for the environment, but also to the health of workers (Sambyal and Sohail, 2015). Due to the increase in various developmental activities in different sectors like industrial, transportation and other related fields, the rising problem of air pollution is increasing day by day.

Respirable dust is a useful indicator of the level and distribution of heavy metals contamination in the atmosphere derived from the natural and anthropogenic activities which may affect air quality including harm to human life (Aybek and Selcuk, 2007). Combustion from burning e-waste creates fine particulates matter, which is linked to pulmonary and cardiovascular disease (Lucy McAllister 2013). The respirable fraction of particulate matter is composed of the very fine dust which is able to reach the lower bronchioles and alveolar regions of the lung, this dust has been implicated to have the potential to carry a high loading of contaminated species such as heavy metals and organic pollutants (Ahmed and Ishiya., 2006).

However, the PM_{10} (thoracic size fraction) and $PM_{2.5}$ (alveolar size fraction) are usually use as the monitoring parameter of air quality. Some of the major sources that emit heavy metals such as As, Mn, Co, Cd, Cu, Ni and Zn into ambient air and its surroundings are domestic, fuel combustion, transportation or mobile emission and industrial processes (Zheng *et al*., 2010, Van *et al*., 2014). Besides these natural sources of heavy metals in the air are mainly from fire in forests and vegetations, volcanoes, dust containing metals getting blown into the atmosphere, which is intensified in dry regions without vegetation coverage.

Most commonly, heavy metals likes Pb, Cu and Zn often exist in the particulate phase of air. According to International Agency of Research on Cancer, most of the heavy metals are harmful to human, and some even have the carcinogenic effect. Pb and Cd known to have the adverse effect on cardiovascular health where as the Cr, Cu and Zn are the important nutrients of food but inhalation of these metals may damaging to the lung (Singh B. R. et al., 2015). Studies revealed that the current level of atmospheric particulates may have serious effect on health, especially in some urban and industrial areas (Hieu and Lee., 2010) due to the presence of heavy metal in it.

Thus, the objective of this study was to determine the concentration and possible source of heavy metals (Pb, Cd, Cu, Zn, Cr and Ni) in Respirable dust (PM_{10}) during summer and monsoon season at all sampling sites. Finding of this study not only provide valuable information on environmental quality of Moradabad, but also present a scientific context of environmental effects of primitive e-waste recycling and industrial activities so that further regulatory and scientific attention can be drawn to the issue.

II. Material And Methods

2.1. Site Description and sampling: For the study, five monitoring stations have been selected based on the predominance of residential, industrial, commercial and vehicular activities existing in the local areas. Location of the study area along with sampling stations and relevant features of sampling stations are given in table 1 and fig. 1 respectively.

Taking the predominant land-use pattern as the selection criteria, PM_{10} samples were simultaneously collected for two different season (summer and monsoon) during one year period from April 2015 - September 2015. The samples were collected with the help of Respirable Dust Samplers (RDS) APM-460 NL (Envirotech, New Delhi) on whatman glass fiber filter paper – GF-A. The instrument was operated at a flow rate of 1.0-1.5 m^3/min and the monitoring of pollutants is carried out for 24 hours (8-hourly sampling for particulate matter) twice a week. Special attention was paid while selecting sampling locations. Priority was given to guidelines prescribed by Central Pollution Control Board of India (CPCB, 2009) along with machine safety and availability of electricity. In the present study, 51 observations were made for summer and monsoon seasons throughout the year.

2.2. Analytical techniques: 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±2oC 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 (5 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°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). To get the final concentration results of the blank samples are subtracted from the exposed samples. To get the mass of each metal, the concentration of metals in the samples is then multiplied by the sample volume (i.e. 25 ml). These values are subsequently divided by corresponding total volume of sampled air to get the concentration of metals in the sampled air.

2.3. Data Analysis Technique: Basic statistical parameters such as mean and standard deviation are computed along with correlation analysis, while co-factor analysis statistics in terms of Principal Component Analysis (PCA) as given by Lee and Hieu, 2011. PCA was conducted by the varimax rotated factor matrix method, based on the orthogonal rotation criterion with Kaiser Normalization.

III. Result And Discussion

3.1. Mass Concentration of Respirable Dust (PM_{10}): The median values with ranges of PM_{10} concentrations that were observed using RDS over the study periods is showing in Fig. 2. The range of mass concentrations varied considerably over time from 70 to 281 μgm^{-3} at SI, 75 to 348 μgm^{-3} at SII, 60 to 250 μgm^{-3} at SIII, 53 to 271 μgm^{-3} at SIV, 40 to 206 μgm^{-3} at SV. It may be attributed to the combination effects of industrial emissions, burning and processing of e-waste, vehicular movement etc. All mean values of PM_{10} concentration are found more than the recommended concentration of NAAQS (100 μgm^{-3}) except SV (40 μgm^{-3}) and SIV (53 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} (50 μgm^{-3}) at all the study area.

The lowest concentration was found at PTC (SV) the residential area which surrounds by greenery while the highest was found of Karula followed by Nagphani which is characterized by e-waste processing with small number of industrial activity and soil dust. The higher PM_{10} concentrations at industrial sites i.e. Peetal Nagri and Mughalpura may reflect a significant contribution of stationary industries, contribution of mobile sources and anthropogenic sources compared to the other sites.

3.2. Impact of meteorological condition and seasonal variation on PM_{10} concentration: The mean concentrations of PM_{10} in summer were higher than those in monsoon (table 2). Similar trends were reported for particulate matter from different areas of India such as Jharia coalfield (Ghose and Majee, 2000), Sambalpur region, (Chaulya, 2004), Raniganj–Asansol area (Reddy and Ruj, 2003) and around Dhanbad city (Jain and Saxena, 2002). In general, high concentrations of PM_{10} in summer are blamed to the effect of dust storm and high wind velocity causing dispersal of pollutants in this season (Mahima *et al*., 2013). Even after the main streams of dust events passed through the region of interest, there were remaining fine particles which contributed to increased local PM concentrations. During the summer sampling period the ambient air temperature at the sampling site was lower than that during the monsoon ones (table 3).

The levels of PM_{10} can also be influenced by other factors like meteorology and long- range transport in the troposphere. In addition to emissions from the local sources, the variations in meteorological parameters were observed to play important role in the levels of pollutants in many urban areas of the world (Verma *et al*., 2010). Among the several meteorological parameters, the local wind can significantly influence the concentrations of various pollutants in e-waste burning and industrial sites. The seasonal variation was found as higher concentrations for PM_{10} occurred in summer period (April-June) at all the study area than those of monsoon (July- September) (fig. 3). The concentration of PM_{10} at SI (187 and 178 μgm^{-3}), S II (200 and 195 μgm^{-3}), SIII (174 and 162 μgm^{-3}), SIV (149 and 132 μgm^{-3}) and S V (126 and 113 μgm^{-3}) was found during the summer and monsoon season respectively.

Monsoon seasons in Moradabad usually has good amount of precipitation and high relative humidity (table 3). So these meteorological conditions, such as increased rainfall and humidity, during the monsoon periods can greatly decrease PM concentrations in ambient air via rainout or washout mechanisms (Tripathi *et al*., 2011). Thus the size of PM_{10} exposed to high humidity or rain drops may increase removal processes of the particles, easily. Thus, the difference in PM_{10} concentrations between the summer and the monsoon periods can be explained by the difference in weather pattern or meteorological condition for each specific season.

3.3. Heavy metal concentrations (μgm^{-3}) in PM_{10}: Mean concentrations of heavy metal at all sites along with standard deviation were presented in table 4 while the mean concentration of heavy metals during summer and monsoon season is represented in fig 4. The mean lead concentration at all the sites was found higher from the permissible limit (i.e. 1.5 μgm^{-3} of National Ambient Air Quality Standard, 2009) during both (summer and monsoon) season. The maximum mean concentration of Pb was found at Karula (5.93 μgm^{-3}) during summer and minimum at P.T.C. (0.96 μgm^{-3}) during monsoon. The higher concentration of Pb is mainly due to traffic volume (Xia and Gao, 2011) because lead pollution due to leaded gasoline still occurs in few cities (Prajapati *et al*., 2009; Andra *et al*., 2011). The mean concentration of Cd was also found higher than the permissible limit i.e. 0.015 μgm^{-3} at all the sites during both seasons except P.T.C. (0.006 μgm^{-3}) during monsoon season. The maximum concentration of Cd was found at Nagphani during summer season i.e. 4.82 μgm^{-3}. Extremely high concentrations of metals such as Pb, Cd are basically due to the vehicular activities and burning of printed circuit boards (PCBs) in Moradabad, as about some estimation 50 percent of PCB used in appliances in India end up in Moradabad. Workers process it to recover metals such as copper, aluminum and gold.

The maximum concentration of Cu and Zn was found at found at Peetal Nagri (61.62 and 94.24 μgm^{-3} respectively) and the minimum was found at P.T.C. (7.77 and 13.06 μgm-3 respectively) during the monsoon. Peetal nagri was surrounded by many small and large scale brassware industries and here Brass (60% Cu and 40% Zn) and German silver (55% Cu, 35% Zn and 10% Ni) are the main alloys used for molding purpose in making brassware items and other utensils in Moradabad. So the major causes of high concentration of these metals are brassware industries which are specialized in cutting, grinding, scraping, polishing etc. (Pal *et al.*, 2014). Besides this, Peetal Nagri (industrial site), is a major exporting centre of brasswares which is situated along the major road connected to Delhi, so the vehicular traffic as well as industrial activity could be the major source of Zn, Cu and Cr.

The maximum concentration of Cr and Ni was found at Nagphani (4.35 and 5.01 μgm^{-3} respectively) during summer while minimum was found at P.T.C. (0.07 and .0052 μgm^{-3} respectively) during monsoon. The mean concentration of Ni was found below the permissible limit (2.5 μgm^{-3}) at all the sites during both seasons except Nagphani and Karula. Dismantling, heating, open air burning of e-waste etc. are the sources of Ni and Cr. As in e-waste Ni was used in alloys, batteries, relays, semiconductors while Cr was found in steel as corrosion protection, data tapes, circuit boards, photocopying-machines (printer drums). The emission of toxic metal i.e. Ni, Cd, Pb, Cr, Zn, Hg, Co and As in the surrounding air by burning of e-waste affects the lungs, kidney, reproductive & nervous system, kidney and cause cancer (Leung *et al*., 2008).

The average concentration of all heavy metal at Mughalpura was found lower than the other sites except the residential area i.e. P.T.C but it have also a significant concentration of metals like Pb, Zn and Cu due to the presence of brassware industries.

It was interesting to note that not much variation was found in the concentration of heavy metals during both seasons. Similar finding were reported by Gajghate and Hasan (1999).

3.4. Correlation coefficient of heavy metal concentration in PM_{10}: The Pearson's correlation coefficient (r) was calculated from the elemental concentration in order to predict the possibility of a common source of PM_{10} (Table 5). The significant positve correlation was found between Zn-Cu (r=0.984 and 0.953), Cd with Cr (r=0.921 and r=0.935) and Cu (r=0.832 and r=0.795), Pb with Cd (r=0.806 and 0.876) and Cr (r=0.863 and r=.808) in both summer and monsoon seasons, respectively. Similarly, significant positive correlation were found between Ni with Cr (r=0.847 and r=0.822) and Cd (r=0.822 and r=0.746) during summer and monsoon season respectively. It may be due to the industrial and anthropogenic activities like vehicular emission and burning of e-waste. There were also found a moderate correlation between Cu-Pb (r=0.499 and r=0.638), Cr with Cu (r=0.611 and r=0.719) and Zn (r=0.568 and r=0537) in both summer and monsoon season. Based on the correlation study, it may be concluded that Zn, Cu, Cd, Pb and Ni were contributed by some common sources, probably by burning of e-waste, industrial, vehicular and anthropogenic sources.

3.5. Factor Analysis: The principal application of factor analysis is to reduce the number of variables. This method focuses on cleaning up the factors. PCA (principal component analysis) was applied to determine the correlation between pollutants and to identify the source profile of heavy metals in PM_{10}. Table 5 shows the results of PCA that was performed to identify common sources of heavy metals with their variance in PM_{10} during summer and monsoon sampling time. PCA results of trace metals in PM_{10} for summer showed three factors accounting for 96.912 % of the overall variance. Factor 1 had high loadings for Pb, Cr and Ni which explained 51.386 % of the total variance. This factor is associated with burning of PCB and e-waste (Pb, Cr, Ni) and traffic emission sources (Pb, Cr) (Chang *et al.*, 2009). Thus, factor 1 can be identified as mixed sources of e-waste processing activities (such as burning and heating of PCB, dismantling and acid bath) and traffic emissions. Factor 2 explained 42.13% of the total variance and correlated with high loadings on Cu and Zn. This factor represented the use of Cu and Zn for molding purpose in making brassware items and other utensils. Another possible source of Zn and Cu is road traffic (diesel engine and wearing of brakes). Factor 3 explained 3.440% of the total variance and characterized by Cr, mainly derived from the anthropogenic activities.

In monsoon, three factors obtained in the PCA analysis of heavy metals in PM10 accounted for 98.06% of the overall variance. Factor 1 (37.82%) had high loadings for Pb and Cd which can be the traffic emission/road dust and burning of PCB. Chang *et al.* (2009) reported that road dust near a industrial area had high loadings on Zn and Pb. Factor 2 (34.54%) with very high loadings on Cu and Zn could be assigned as industrial emissions because the main sources of these metals in the PM are cutting, grinding, scraping etc. of the brassware products. Factor 3 (25.63) had high loading for Cr which is originated by anthropogenic activities.

IV. Conclusion

Assessment of air quality monitoring data around e-waste burning and industrial areas showed significant spatial and seasonal variations in concentrations of PM_{10} and their heavy metal content depending on pollutant emission or formation and pollutant dispersion mechanisms, which are also influenced by meteorological conditions. Wind speed, relative humidity, temperature and rainfall were the governing parameters of seasonal variations in air pollutant concentrations in the area. Overall site specific assessment of PM_{10} data represent that Karula is the most polluted area in terms of dust loading with a maximum mean concentration of 200 and 195 μgm^{-3} followed by Nagphani i.e. 187 and 178 μgm^{-3} during summer and monsoon season respectively. The concentration of PM_{10} in summer was found higher than monsoon. The PC I for summer (51.386%) is characterized by Pb, Cr and Ni which represents burning of PCB (e-waste) and traffic emission while for monsoon (37.82%) is characterized by Pb and Cd which represents road dust and burning of PCB. The PC II for summer and monsoon season i.e. 42.13% and 34.54% respectively is associated with Industrial activities along with road traffic (i.e. Cu and Zn). The PC III for summer and monsoon season i.e. 3.44% and 25.63% respectively is identified as anthropogenic sources and characterized by Cr. These results are also supported by correlation study. So e-waste burning, industrial activities, wind-blown dust, vehicular emission as well as anthropogenic activities are the major causes of air quality deterioration of Moradabad. The outcomes of this study may provide a comprehensive catalogue for outlining a proper scheme for required mitigative or preventive measures are the other causes of air quality.

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Table 1: Relevant features of air quality monitoring stations

Area	Sampling Location	Activity
E-waste burning and processing area	• Nagphani (SI) • Karula (SII)	Burning of PCBs to extract metal, hammering, dismantling and segregation of components from PCBs, acid bath, direct heating by blow torch, open air burning, burning of solid waste, commercial and residential activity.
Industrial area	• Peetal Nagri (SIII) • Mughalpura (SIV)	Cutting. Grinding, electroplating and polishing of brassware items, metallurgical activity, commercial activities, vehicular movement on paved road and unpaved road, fossil fuel burning and residential activities etc.
Residential area (Control Site)	• P.T.C. (SV)	residential activities, commercial activities, vehicular movement, litter burning etc.

Table 2: Mean concentration of respirable dust (PM_{10}) during summer and monsoon season

Sites	Summer			Monsoon		
	Mean±S.D.	**Max.**	**Min**	**Mean±S.D.**	**Max.**	**Min**
S I	187±10.53	270	70	178±6.02	289	75
S II	200±3.05	348	75	195±5.50	300	87
S III	174±9.84	350	60	162±23.71	280	89
S IV	149±24.54	271	84	132±11.93	201	53
S V	126±11.26	206	70	113±8.02	183	40

Table 3: Meteorological conditions at the sampling site during sampling periods

Meteorological parameter	Summer	Monsoon
Temperature (^{O}C)	35.71±10.42	36.11±7.07
Humidity (RH) (%)	34.56±20.52	68.05±17.48
Wind speed (km/h)	8.47±0.89	7.09±1.88
Rainfall (mm)	1.89±3.41	10.91±10.61

Table 4: Mean concentration of heavy metal at different sites of Moradabad

Sites	**Season**	**Pb**	**Cd**	**Cu**	**Zn**	**Cr**	**Ni**
Nagphani	**S**	4.67±0.460	4.82±0.180	50.68±1.440	90.47±4.099	4.35±0.563	5.01±0.195
	M	3.61±0.458	3.53±0.404	43.97±1.781	81.71±3.835	3.03±0.873	4.74±0.490
Karula	**S**	5.93±0.152	4.36±0.450	37.98±1.568	58.55±0.661	4.11±0.351	4.15±0.925
	M	5.33±0.378	4.6±0.8	34.09±2.656	50.67±1.910	3.41±0.125	2.15±0.313
Peetal Nagri	**S**	2.03±0.472	2.63±0.265	61.62±0.358	94.24±4.665	1.07±0.110	1.98±0.958
	M	2.99±0.457	2.26±0.277	40.12±4.06	90.08±3.033	1.023±0.150	1.53±0.452
Mughalpura	**S**	2.87±0.121	2.22±0.092	20.31±0.975	23.31±2.766	0.34±0.29	2.43±0.043
	M	2.9±0.435	0.53±0.409	19.5±2.365	27.32±3.500	0.21±0.31	0.2±0.060
P.T.C.	**S**	0.96±0.14	0.007±0.0015	7.77±0.469	13.06±1.006	0.94±0.280	0.0058±0.0012
	M	0.53±0.102	0.006±0.001	8.23±0.680	10.36±0.723	0.07±0.011	0.0052±0.001

S= Summer, M= Monsoon

Table 5: Correlation coefficient between the concentration values of analyzed heavy metal in PM_{10} at different site during summer and monsoon season.

Metal	**Pb**	**Cd**	**Cu**	**Zn**	**Cr**	**Ni**
Pb	1	0.806	0.499	0.395	0.863	0.863
Cd	0.875	1	0.832	0.802	0.921	0.822
Cu	0.683	0.795	1	0.984	0.611	0.693
Zn	0.504	0.633	0.953	1	0.568	0.603
Cr	0.808	0.935	0.719	0.537	1	0.847
Ni	0.526	0.746	0.831	0.729	.822	1

Summer n= 25 , **Monsoon** n= 26

Table 6: Rotated factor loading of heavy metals in PM_{10}

Variable	**Summer**			**Monsoon**		
	Factor 1	**Factor 2**	**Factor 3**	**Factor 1**	**Factor 2**	**Factor 3**
Pb	.950	.171	.139	.935	.280	.151
Cd	.671	.625	.388	.783	.358	.466
Cu	.337	.937	.051	.423	.824	.368
Zn	.220	.965	.134	.226	.938	.247
Cr	.816	.341	.439	.699	.219	.668
Ni	.880	.438	-.097	.259	.510	.809
%Variance	51.336	42.137	3.440	37.82	34.546	25.630
Cumulative%	51.336	93.473	96.912	37.82	72.439	98.069
Sources	e-waste processing and traffic emission	Industrial activities (molding Purpose) and road traffic	Anthropogenic activities	Burning of PCB and traffic emission/road dust	Industrial emission (grinding, scrapping etc.)	Anthropogenic activities

Fig. 1: Location of the study area along with sampling station

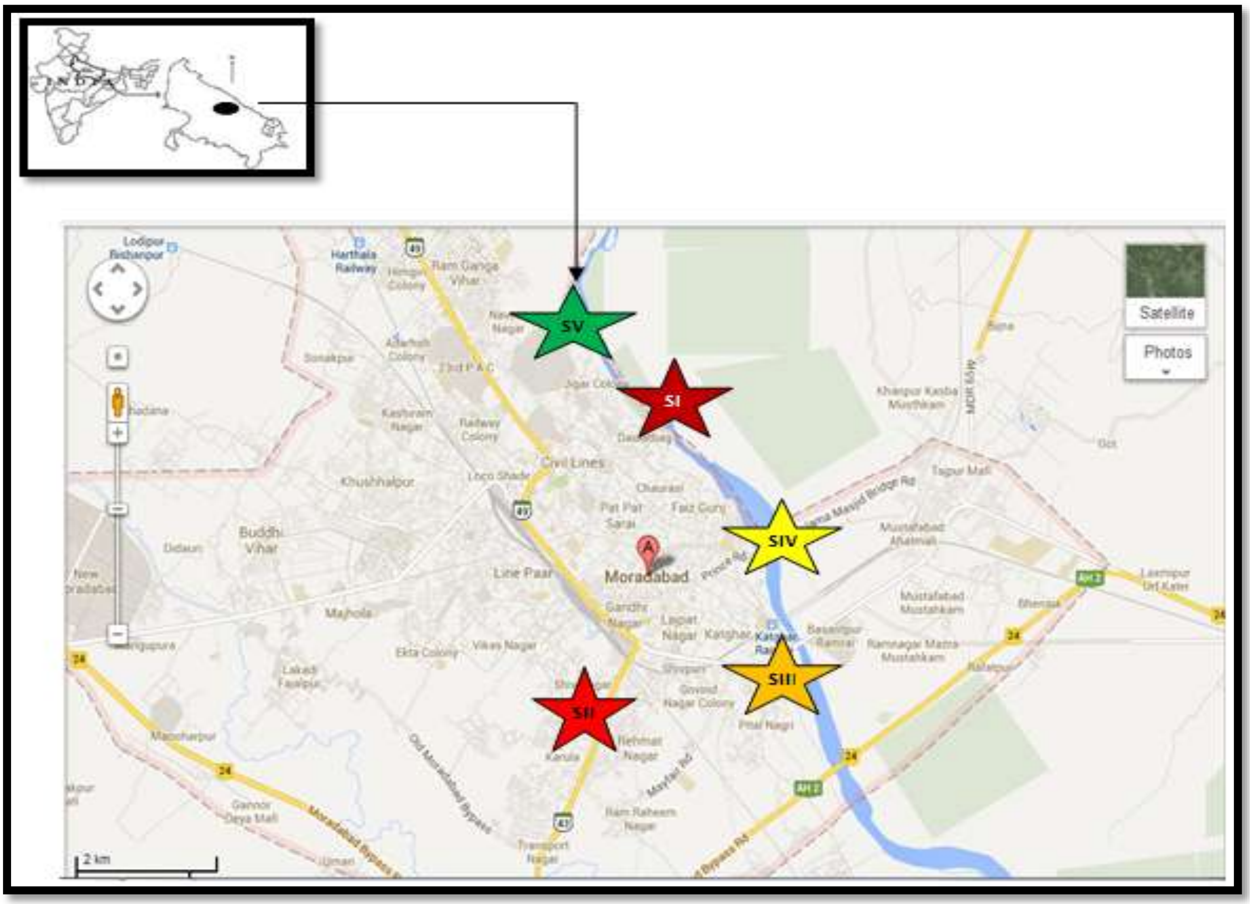

Fig. 2: PM_{10} concentration distribution in summer and monsoon

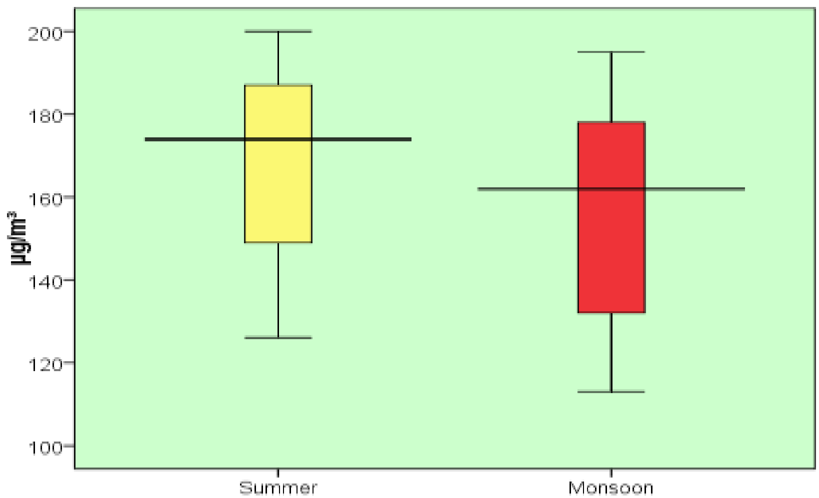

Fig. 4: Seasonal variation in PM_{10} concentration.

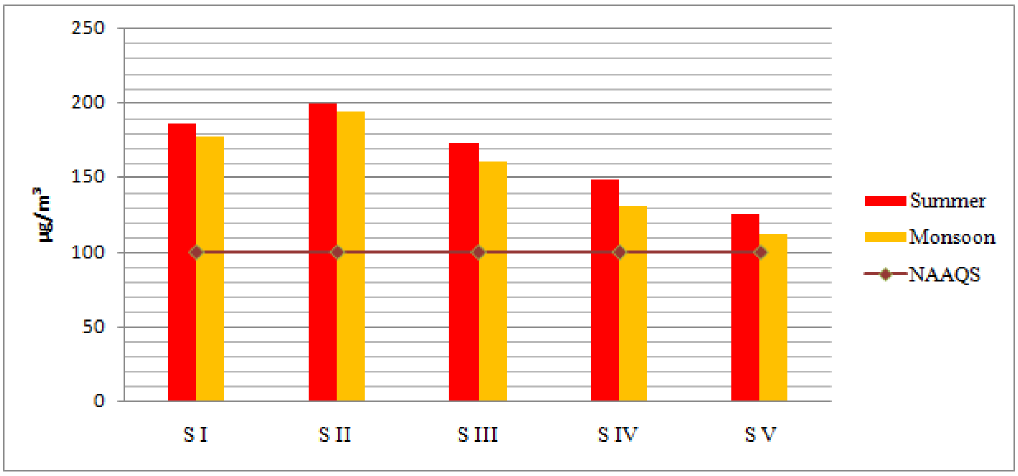

Fig. 5: Concentration of heavy metals (Pb, Cd, Cu, Zn, Cr and Ni) at different sites of Moradabad during Summer and Monsoon