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Trace element composition of $PM_{2.5}$ and PM_{10} from Kolkata – a heavily polluted Indian metropolis

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ABSTRACT

Elemental composition of $PM_{2.5}$ and PM_{10} was measured from 16 locations in Greater Kolkata in Eastern India. Sampling was carried out in the winter months of 2013–2014. $PM_{2.5}$ and PM_{10} mass concentrations ranged from 83–783 μg/m^3 and 167–928 μg/m^3 respectively. 20 elements were measured with an Agilent 7700 series ICP-MS equipped with a 3rd generation He reaction/collision cell following closed vessel microwave digestion. In both size fractions Fe, Na, Al, K, Ca were present in high concentrations (>1 000 ng/m^3), Mn, Zn and Pb demonstrated medium concentrations (>100 ng/m^3), and Sc, V, Co, Ni, Mo, Cd, Sn and Sb had low concentrations (<100 ng/m^3). Ca, Al, Mg, Sc, Ti, Mn and Fe were concentrated in the PM_{10} fraction, while the toxic metals (Cr, Ni, Zn, Mo, Sn, Sb, V, Co, Cu, Cd and Pb) were concentrated in the $PM_{2.5}$ fraction. Al normalized Enrichment Factors (EF) showed EF<10 for Ti, Mg, Sc, Fe, Mn, Na, K, Ca, V, Co which is indicative of crustal sources, 100>EF>10 for Ni, Cr, Cu is possibly industrial influence and 1 000>EF>100 for Sn, Zn, Mo, Sb, Pb, Cd is related to industrial, high temperature combustion and vehicle sources. Factor analysis identified three possible sources for PM_{10}; (1) abraded vehicular part related road dust, exhaust gases of car and municipal waste incineration (2) industrial emissions, and (3) coal combustion and non–ferrous metal smelting and three possible sources for $PM_{2.5}$; (1) abraded vehicular part related road dust and industrial emissions (2) exhaust gases of cars and municipal waste incineration, and (3) coal combustion and non–ferrous metal smelting. In a risk evaluation using a U.S. EPA IRIS, chromium was found to have the highest excess cancer risk.

Keywords: *Particulate matter, heavy metals, enrichment factor, factor analysis, excess cancer risk*

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1. Introduction

Trace element composition of the respirable size fraction of particulate matter (PM) is being increasingly monitored since several metals have been implicated in adverse human health outcomes (Freitas et al., 2010; Garcia et al., 2011). Recent epidemiological studies reveal that the heavy metals in the air borne particles are of major concern due to their numerous health effects (Andersen et al., 2006; Sarnat et al., 2006; Liu et al., 2009; Mavroidis and Chaloulakou, 2010). The PM_{10} and $PM_{2.5}$ are of particular concern to human health as they are in the inhalable and respirable size ranges, respectively. These particles are readily trapped by the alveoli of the lungs (Perez et al., 2008). Not only do the toxic heavy metals enter our body through the respirable particulate matter (RPM), the air particulates can cause coughing, wheezing, shortness of breath, aggravated asthma and lung damage leading to premature death (Dockery et al., 1993; Dockery and Pope, 1994). Air pollution plays an important role in the genesis and augmentation of allergic disorder and it is described as a disease of civilized society (Ownby, 1990; Bonini et al., 1994). There exists a strong link between air pollution and cardiovascular disease and cancer (WHO, 2014). Hence, with increased industrialization it has become important to know the ambient atmospheric composition and have a better understanding of the emission sources.

Indoor and outdoor air pollution was responsible for an estimated 7 million deaths in 2012 (WHO, 2014), and is the world's single largest environmental health threat. Air quality has become a major concern in many of the Southeast Asian countries which is home to more than 50% of the global population. In the developing nations of Asia, the main sources of air pollution are diesel soot from cars and trucks, emissions from industries and power plants, municipal solid waste incineration, biomass burning and the dust from endless urban constructions (Hien et al., 2001; Shridhar et al., 2010; Shah et al., 2012). The urban populations of India are exposed to some of the highest pollutant levels in the world (WHO, 1999; Smith et al., 2000). A blood sample study of 1 852 children in 5 major cities in India reported 51.4% had levels of lead in their blood above 10 μg/dL (The George Foundation Report, 1999). Twenty three cities in India with populations more than a million have air pollution levels exceeding WHO standards (Chowdhury, 2004). Respiratory disease related problems caused by both indoor and outdoor pollution in Indians have been identified in Indian cities like Delhi, Kolkata, Mumbai, Lucknow and Ahmedabad (Chowdhury, 2004).

The latest report by the Central Pollution Control Board of India (CPCB, 2012) shows that Delhi, the national capital and Kolkata (erstwhile Calcutta), capital of West Bengal and financial hub of Eastern India are among the worst affected cities in terms of air pollution. The PM concentration on 8 hour exposure basis was recorded to be 2 118 μg/m^3 from an industrial site in Delhi and

490 μg/m^3 was recorded from a university campus at the heart of the national capital (Kushwaha et al., 2012). Indian Council of Medical Research (ICMR) vindicated air pollution as one of the lung cancer causing agents and placed it in the same category as tobacco, UV radiation and plutonium. Kolkata is the second largest metropolis in South Asia and is one of the worst polluted cities in the world. The Greater Kolkata covering an area of 1 480 square km is home to numerous large and small–scale industries of different categories, thermal power plants, port and is notorious for heavy traffic. As a result of heavy air pollution, 41.3% and 47.8% of Kolkata population suffers from upper and lower respiratory tract symptoms respectively, compared to 13.5% and 35% of rural Bengal population (WBPCB, 2001). Suspended particulate matter alone was responsible for more than 10 000 premature deaths in Kolkata in 1995 (Kazimuddin and Banerjee, 2000).

Previous studies on Kolkata air quality showed that the main sources of air pollution were solid waste dumping, vehicular emissions, coal combustion, cooking and soil dust at a residential site and vehicular emissions, coal combustion, electroplating industries, tyre wear and secondary aerosols at an industrial site (Karar and Gupta, 2007). A study conducted at major traffic junctions of Kolkata showed that the key pollutants like lead, NO_X, PM_{10}, SO_2, CO are in excess of permitted levels (Ghose et. al., 2004). The aim of this study was to quantify the levels of toxic metals in PM_{10} and $PM_{2.5}$ in and around Kolkata. Elemental compositions are also used for quantitative apportionment of the sources. We set up our collecting stations in residential areas close to traffic junctions, coal fired power plants, industrial belts, waste incineration plants, cement factories and brick kilns in and around Kolkata. The sites were carefully selected to avoid undue influence of direct emissions coming from heavy city traffic or industrial work, yet they were located within residential areas or open fields close to the emission sources thus enabling us to capture the cities' emissions throughout the daytime. In Indian metropolis there are no clear demarcation between residential areas and industrial belts. People live close to traffic junctions, industrial estates, power plants etc., and commute vast distances across the city and suburbs for their livelihood. Hence the purpose of this study was to investigate the elemental composition of PM_{10} and $PM_{2.5}$ of the Greater Kolkata area keeping in mind the exposure a working person might have on a daily basis. For this purpose, we sampled from multiple locations in and around Kolkata to obtain an accurate representation of the average air quality of the city during the winter time when pollution levels are the highest.

2. Materials and Methods

2.1. Study area

Kolkata (22°32'N, 88°22'E), the capital of the Indian state of West Bengal, is the 3rd most populous city of India according to a 2011 census. The city and its suburbs are home to approximately 14.1 million people. Though the city is the economic, cultural and educational hub of eastern India it is facing a tremendous burden of traffic congestion, poverty, overpopulation, and logistic problems related to the population burst. Kolkata has a tropical wet–and–dry climate with annual mean temperature of 26.8 °C. The city experiences 3 distinct seasons, summer, monsoon and winter. May is the hottest month with daily temperatures ranging from 27–38 °C. South–West Monsoons pick up moisture from the Bay of Bengal and bring rain from mid–June to end of September. Winter lasts for about two–and–a–half months, with January being the coldest month with temperatures varying from 9–23 °C (Spiroska et al., 2011). Sampling was carried out in December 2013 and January 2014. The winter months were chosen because the inversion layer remains close to the ground and pollutant levels are highest during the cooler months.

2.2. Sampling

Two Deployable Particulate Sampler (DPS) pumps (Leland Legacy) with a pumping efficiency of 10 L/min were used to collect the PM_{10} and $PM_{2.5}$ simultaneously. The PM was collected on 47 mm quartz filters. Sampling stations were selected at 16 different locations in and around Kolkata (Figure 1). Brief descriptions of the locations with their GPS coordinates are listed in Table S1 (see the Supporting Material, SM). During our sampling period the daily average temperature varied between 17 °C to 22 °C and the predominant wind direction was N–NNW.

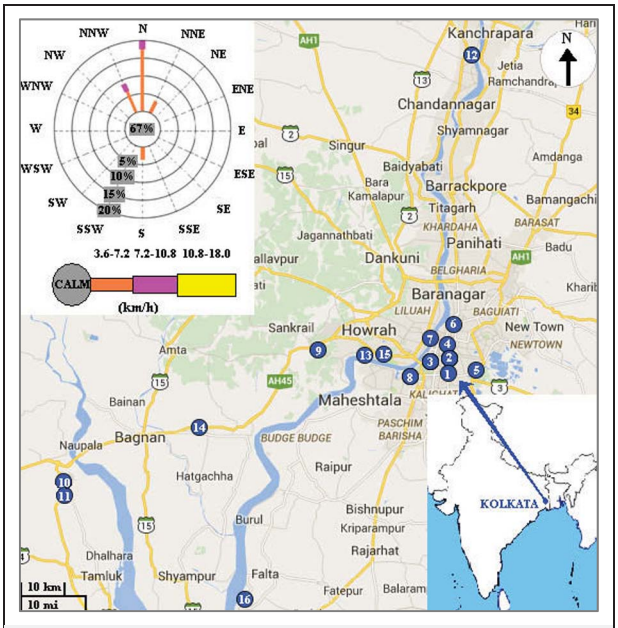

Figure 1. *Map showing the sampling sites and wind rose showing the prevalent wind direction in Kolkata during the study period (December 2013–January 2014).*

Quartz filters were dried overnight at 400 °C in an oven, weighed and packed in Millipore petri slides with a cover to prevent any contamination. Sampling was performed between 6 AM and 6 PM. The working population of the metropolis is exposed to maximum outdoor pollution during this 12 hour period. After sampling, filters were immediately sealed in the petri slides and stored in refrigerator.

2.3. Chemical analysis

Before acid digestion the filters were weighed to calculate the concentration of the particulate matter. A programmable Milestone microwave system (ETOS EZ, Italy) was used to digest and prepare samples for ICP analysis. Twenty elements were measured with an Agilent 7700 series ICPMS equipped with a 3rd generation He reaction/collision cell. Details of the chemical analysis and instrument parameters are listed in the SM (Text S1).

2.4. Enrichment factor calculation

Calculation of enrichment factors (EF) is often used to detect contributions of non–crustal sources on observed concentrations of elements. Calculations are generally based on the average upper continental crustal composition given by Rudnick and Gao (2003). The EF of an element in a PM sample is defined as:

$$EF = \left(\frac{\left(\frac{X}{Ref}\right)_{Sample}}{\left(\frac{X}{Ref}\right)_{Crust}} \right) \quad (1)$$

where *X* and *Ref* are respectively the element and the reference element concentration in the sample and crust. Aluminum and iron are the most common elements used for this purpose although there are no strict requirements for which reference elements to use. In this work, both Al and Fe are present in high concentrations. Since sampling sites are located nearby traffic junctions and industries where road traffic and industrial sources can be a potential source of Fe, Al was chosen as reference element. Figure 2 shows the pattern of EF for PM_{10} and $PM_{2.5}$.

2.5. Statistical analysis of the data

Exploratory factor analysis was conducted separately on two sets of data corresponding to PM_{10} and $PM_{2.5}$. In each case, factorability of the correlation matrix was established by the fact that there were many correlations in excess of 0.3 (Tabachnick and Fidell, 2014). Factors were extracted using the *factanal* function of the *stats* package of *R*, which uses the maximum likelihood factoring technique. Both varimax (orthogonal) and promax (oblique) rotation during initial analysis were tested, and the adequacy of rotation was examined by observing scatter plots with pairs of rotated factors as axes and variables as points. Varimax rotation was selected for the final analysis due to its simplicity and ease of interpretation.

2.6. Determination of excess cancer risk

In this study, excess cancer risks (ECRs) were calculated for concentrations of $PM_{2.5}$ bound metals. ECR was calculated by using the formula: Excess cancer risk (inhalation) = concentration of pollutant (μg/m^3) × unit risk (μg/m^3) (U.S. EPA, 1989). Carcinogenic group and unit risks of the metals are provided by U.S. EPA IRIS (Integrated Risk Information System) (U.S. EPA, 1998). Regarding bioavailability of the metals, we assumed that the $PM_{2.5}$ bound heavy metals represents the total ambient concentrations of the metals and 100% of the metals were absorbed into the body by lungs.

3. Results and Discussion

3.1. Particulate matter

The average PM_{10} and $PM_{2.5}$ concentrations were 445± 210 μg/m^3 (range: 167 to 928 μg/m^3) and 313±181 μg/m^3 (range: 83 to 783 μg/m^3), respectively. This is 4.45 and 5.2 times higher than the daily average concentration of PM_{10} and $PM_{2.5}$ prescribed by National Ambient Air Quality Standards (NAAQS, 2009). The highest PM_{10} and $PM_{2.5}$ concentrations were measured in the traffic junctions (Site 1 and Site 2). Site 13 close to brick kilns also recorded high values of PM_{10} (611 μg/m^3) and $PM_{2.5}$ (422 μg/m^3). Previous studies in residential and industrial areas of the city reported an annual average PM_{10} concentration of 140±43 μg/m^3 and 197±88 μg/m^3 respectively (Karar and Gupta, 2007). Another study carried out in industrial, commercial and residential areas in Kolkata reported PM_{10} concentration of 400 μg/m^3, 350 μg/m^3, and 300 μg/m^3 respectively in the months of December 2004 to February 2005 (Spiroska et al., 2011). Measurement of fine particle air pollution ($PM_{2.5}$) in Kolkata and Delhi showed an average concentration of 305 μg/m^3 and 230 μg/m^3 respectively during the winter (Chowdhury, 2004). Several studies also showed that the maximum average PM concentrations occurred during winter and the minimum average concentrations occurred during the summer, a feature consistent with most of the Indian cities (Chowdhury, 2004; Spiroska et al., 2011; WBPCB, 2012). However all the previous studies had their sampling location at one or two stations whereas in the present study the stations are in multiple locations. All the locations in the present study showed PM_{10} and $PM_{2.5}$ values higher than NAAQS of 100 μg/m^3 and 60 μg/m^3 respectively. The high levels of PM in the city are due to vehicular traffic, industrial emissions, road and soil dust, ongoing constructional activities, municipal wastes incinerations and other industrial activities.

3.2. Trace elements

Twenty elements were analyzed in the air of the Kolkata metropolitan area in the PM_{10} and $PM_{2.5}$ fractions (Table 1). There are three distinct concentration groups for the mean concentration of metals in PM_{10}. Fe has the highest concentration (11 242 ng/m^3) with 72% of the metal concentrating in the PM_{10} fraction. In urban air, iron can come from crustal dust, vehicular emissions, construction activities and industrial emissions. The light metal group (atomic number 20 and lower) including Na, K, Ca, Al, Mg were found in very high concentrations (>1 000 ng/m^3) in PM_{10}. Sc was present in trace quantity as it has a very low crustal abundance. The heavy metals (atomic number >20) were distributed in two groups: Zn, Pb, Ti, Mn, Cu and Cr with concentration ranging from 100 ng/m^3 to 761 ng/m^3. The other heavy metals (Ni, Sn, V, Mo, Sb, Cd and Co) were present in trace quantities (less than 50 ng/m^3). The elemental composition of the $PM_{2.5}$ fraction is more important pertaining to human health as PM with diameters less than 4 μm can penetrate into the trachea, bronchi and the alveoli (Perez et al., 2008). In the present study, elements like Ca, Al, Mg, Sc, Ti, Mn and Fe are concentrated mostly in the PM_{10} fraction. However, the alkali metals like Na and K concentrate more in the $PM_{2.5}$ fraction. The heavy metals that are detrimental to human health and are usually attributed to anthropogenic sources are concentrated in the $PM_{2.5}$ fraction. More than 70% of Cr, Ni, Zn, Mo, Sn, Sb, and Pb are in the fine breathable fraction of $PM_{2.5}$. More than 50% of V, Mn, Co, Cu and Cd are also in the $PM_{2.5}$ fraction.

In Table 2 we report the metal concentrations observed in this study and those reported from India and other parts of the world. In all the studies Fe has the highest concentration. Fe concentration in our study is comparable to concentrations measured in suburban/residential Delhi (Khillare and Sarkar, 2012). In spite of having a high concentration the source of iron is concluded to be crustal in Delhi (Khillare and Sarkar, 2012). Cd concentration of the present study is comparable with all the other studies from around the world except for Lahore (von Schneidemesser et al., 2010) which has ~10 times higher Cd levels than our study. Pb concentration of our study is similar to those observed in Delhi and Beijing (Perrino et al., 2011; Duan et. al., 2012; Khillare and Sarkar, 2012) but an order of magnitude lower than Agra and Lahore and an order of magnitude higher than developed countries (Manalis et al., 2005; Jorquera, 2009; Terzi et al., 2010; Theodosi et al., 2010). Sb concentration measured in the present study is less than those measured in Beijing, China (Duan et. al., 2012) and Thessaloniki, Greece (Terzi et al., 2010). Other metals like Cr, Cu, Mn, Ni, Zn concentrations in the present study are comparable or sometimes less than those measured in Delhi, Agra, Beijing and Lahore, however they are definitely more than those measured in developed western countries sometimes by order(s) of magnitude (except for Cu concentration in Tocopilla, Chile due to influence of a nearby Cu mine).

3.3. Enrichment factors

The enrichment factor can indicate natural emissions, marine sources and anthropogenic activities. Elements with EF less than 10 are assumed to be originated from crustal erosion. In the present study, some potentially toxic elements such as Fe, Mn, V and Co are found in this category (Figure 2). Elements with EF values ranging from 10 to 100 are likely to be moderately enriched by anthropogenic sources. Elements such as Ni, Cr and Cu fit into this category and are commonly enriched by industries nearby sampling locations. Highly enriched elements (Sn, Zn, Mo, Sb, Pb and Cd) with EF values greater than 100 are most probably related to industrial (chemicals, electronic), high temperature combustion sources (incineration, oil combustion, coal combustion) and vehicular emissions.

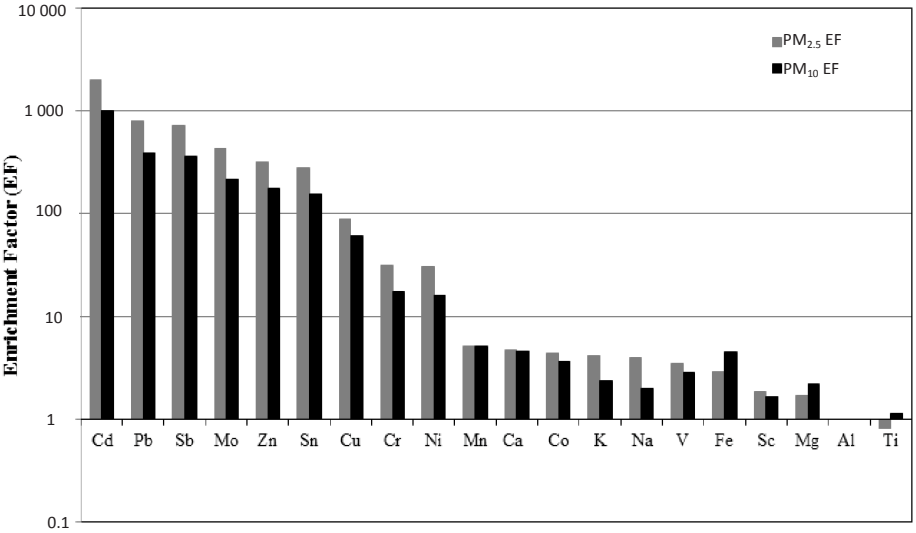

Figure 2. *$PM_{2.5}$ and PM_{10} enrichment factors.*

Table 1. *Statistical analysis of the PM concentrations (μg/m³) and elemental concentrations (ng/m³) of $PM_{2.5}$ and PM_{10} measured during the study period*

N=34	Mean ($PM_{2.5}$)	Min	Max	Mean (PM_{10})	Min	Max	% in $PM_{2.5}$
PM	313±181	783	83	445±210	928	167	70%
Na	2 047±1 434	615	7 425	2 393±1 256	809	8 068	86%
Mg	715±418	214	1 640	2 119±1 346	368	7 066	34%
Al	2 240±2 960	494	14 732	5 223±2 978	877	12 230	43%
K	2 656±1 396	695	7 553	3 486±1 991	1 059	10 198	76%
Ca	3 351±1 912	1 294	8 410	7 610±5 611	1 721	32 811	44%
Sc	0.70±0.50	0.29	2.4	1.49±0.71	0.35	2.95	47%
Ti	85±74	18	357	278±177	44	867	31%
V	9.5±5.2	3.7	27	18±9.3	4.5	42	53%
Cr	84±40	46	192	101±72	42	424	83%
Mn	132±159	15	852	249±227	31	941	53%
Fe	3 150±2 725	1 079	13 236	11 242±15 705	1 448	94 237	28%
Co	2.1±1.4	0.82	7.75	4.1±1.9	1	8.4	51%
Ni	40±18	19	97	48±29	16	171	83%
Cu	58±33	18	185	107±95	21	457	54%
Zn	542±245	158	1 072	761±709	220	4 220	71%
Mo	14±9.3	4.1	41	15±14	3.8	71	93%
Cd	5±3.9	0.82	17	8.6±17	0.9	103	58%
Sn	16±9	6.6	55	21±22	4.1	140	76%
Sb	8.7±7.3	2.7	34	10±8.3	2.2	39	87%
Pb	368±479	37	2 233	394±384	43	1 496	93%

3.4. Factor analysis

We performed factor analysis on 9 metals with enrichment factor greater than 10 (Ni, Cr, Cu, Sn, Zn, Mo, Sb, Pb, Cd) to understand the anthropogenic sources of these metals. To determine the number of factors to be extracted, the set of eigenvalues for each factor and the corresponding scree plot was generated using the *nFactors* package of R. Since eigenvalues less than 1 are negligible from a variance point of view, only factors with eigenvalues ≥1 are included in the results. The number of factors with eigenvalues ≥1 are usually expected to be between the number of variables (9 metals in the present case) divided by 3 and the number of variables divided by 5, which holds for both the cases PM_{10} and $PM_{2.5}$ (Tabachnick and Fidell, 2014). A scree test was used as an additional criterion for deciding on the number of factors. On the scree plot, factors were represented in descending order on the abscissa with their corresponding eigenvalues as the ordinate. The appropriate numbers of factors were determined by identifying the point at which a line drawn through the points on the scree plot changes slope (Tabachnick and Fidell, 2014). Table 3 and Table 4 give the loadings of variables on factors, communalities, eigenvalues, percentage variance, and percentage cumulative variance.

As observed in Table 3, three factors summing to 84.3% of the total variation in the dataset were obtained for PM_{10}. Communalities of all elements in the dataset were in the range 0.62–0.99 indicating that each element was satisfactorily apportioned to the identified factors. Each factor or source was physically interpreted by its association with strongly loaded marker elements, typically emitted from that source.

Table 2. *Comparison of metal concentrations (ng/m^3) in PM_{10} in the present study with those from other parts of India and the world*

Location	Site Type, Sampling Season	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Sb	Zn
This Study	Urban, winter	8.6	101	107	11 242	249	48	394	10	761
Kolkata, India (Karar and Gupta, 2007)	Residential, annual	2	7		87	2	7	40		490
Kolkata, India (Karar and Gupta, 2007)	Industrial, annual	5	6		123	2	8	119		535
Delhi, India (Perrino et al., 2011)	Urban, autumn		39	74	2 500	80	12	430		58
Delhi, India (Khillare and Sarkar, 2012)	Suburban residential, annual	19	230	210	11 200	250	370	460		4 100
Delhi, India (Khillare and Sarkar, 2012)	Urban, annual	8–16	130–200	180–270	8 000–9 600	290–320	280–300	270–410		4 300–4 700
Agra, India (Kulshrestha et al., 2009)	Urban, winter		300	100	2 900	900	200	1 100		500
Beijing, China (Duan et al., 2012)	Urban, winter	4.2	178	55	2 902	113	89	468	18	317.6
Beijing, China (Duan et al., 2012)	Curbside, winter	3.6	22	80	4 480	129	25	324	23	469.5
Lahore, Pakisthan (von Schneidemesser et al., 2010)	Urban, winter	80	30	70	8 200	300	20	4 400		1 100
Tocopilla, Chile (Jorquera, 2009)	Rural, fall season (Nearby Cu mine)			167–1 067	1 332–3 522	22–59	0.9–6.5	12–20		13–33
Istanbul, Turkey (Theodosi et al., 2010)	Urban, annual	1	4	20	700	20	7	70		240
Athens, Greece (Manalis et al., 2005)	Urban, annual	2–3	10–16	13–141		4–21	9–16	25–71		
Thessaloniki, Greece (Terzi et al., 2010)	Urban–industrial, winter	13	17	99	1 010	39	15	39	24	117
Thessaloniki, Greece (Terzi et al., 2010)	Urban–traffic, winter	13	6.64	44	615	30	8.2	24	18	76

Table 3*. Varimax rotated factor matrix for PM_{10}*

Metals	Factor 1	Factor 2	Factor 3	Communality
Cr	0.904	0.064	–0.024	0.823
Ni	0.899	0.042	0.357	0.939
Cu	0.175	0.771	0.249	0.687
Zn	0.140	0.237	0.959	0.995
Mo	0.873	0.057	0.017	0.766
Cd	0.057	0.977	0.090	0.966
Sn	0.076	0.953	0.185	0.947
Sb	0.865	0.258	0.178	0.847
Pb	0.157	0.523	0.564	0.616
Eigenvalue	4.5	2.5	1	
% variance	35.8	31.8	16.7	
Cumulative % variance	35.8	67.6	84.3	

The first factor showed high loadings for Cr, Ni, Mo and Sb (0.9–0.86) explaining 35.8% of the variance. Strong correlation between Cr and Ni (0.81), Sb and Cr (0.80), Sb and Ni (0.84), Sb and Mo (0.8) (see the SM, Table S2) suggest that the characteristics and origin of emissions for all elements may be similar. Individually these elements can have different sources. A five year integrated inventory of atmospheric antimony (Sb) emissions from anthropogenic activities in China showed that the largest contributor of the metal is fossil fuel combustion (61.8%) followed by non–ferrous metals smelting (26.7%) while other sources include municipal solid waste incineration, brake wear and pig iron and steel smelting (Tian et al., 2012). Anthropogenic sources of Cr in the atmosphere include combustion of natural gas, oil and coal, metal industries such as chrome plating and steel production, cement production, incineration of municipal refuse and sewage sludge, and emission from chromium–based automotive catalytic converters (Pacyna and Pacyna, 2001; ATSDR, 2005a). Several studies have identified Ni as a typical tracer for fossil fuel combustion (Artaxo et al., 1999; Pacyna and Pacyna, 2001; Manoli et al., 2002; Khillare et al., 2004). Other sources of Ni include high–temperature metallurgical operations, and municipal waste incineration (Khillare and Sarkar, 2012). Mo is a tracer for fossil fuel combustion (Duan et. al., 2012) and also reported in high concentrations in fly ash generated from municipal solid waste incineration (Haugsten and Gustavson, 2000). After evaluation of the possible sources we interpret the high loadings of Cr, Ni, Mo and Sb to be caused by abraded vehicular part related road dust, exhaust gases of cars (petroleum, diesel combustion) and municipal waste incineration. The second factor explains 31.8% of the variation and presented high loading for Cd, Sn and Cu (0.98–0.78) and moderate loading of Pb (0.52). Cd can be released to the atmosphere by combustion of fossil fuels, iron and steel production, non–ferrous metals production and municipal solid waste combustion (Bernhard et al., 2005; Cook and Morrow, 1995; Chandler, 1996). Anthropogenic sources of Sn includes smelting and refining processes, industrial uses of tin, waste incineration, and burning of fossil fuels (ATSDR, 2005b). Cu can be released to the atmosphere from copper smelters and ore processing facilities. Hence we interpret the second factor to represent industrial

emissions, especially, the metallurgical/electroplating units located in the industrial area of the city (Wang et al., 2001; Ragosta et al., 2002; Quiterio et al., 2004; Zereini et al., 2005; Shah and Shaheen, 2008). The third factor explains 16.7% of the variation and has strong loading of zinc (0.96) and moderate loading of Pb (0.56). Both Zn and Pb can be emitted to the atmosphere from coal combustion and ferrous and nonferrous smelters (Lee et al., 1999; Callender and Rice, 2000; Pacyna and Pacyna, 2001). Hence we interpret the third factor to represent coal combustion (Guerzoni et al., 2005) and non–ferrous smelting (Ho and Lee, 2002).

***Table 4**. Varimax rotated factor matrix for $PM_{2.5}$*

Metals	Factor 1	Factor 2	Factor 3	Communality
Cr	–0.007	0.991	–0.041	0.984
Ni	–0.004	0.920	0.204	0.889
Cu	0.903	0.053	0.080	0.824
Zn	0.874	0.066	0.033	0.769
Mo	0.259	0.708	0.057	0.571
Cd	0.126	0.124	0.979	0.990
Sn	0.838	0.111	0.116	0.728
Sb	0.588	0.537	0.145	0.654
Pb	0.781	0.107	0.611	0.995
Eigenvalue	4.4	2.3	1.1	
% variance	36.9	29.6	15.8	
Cumulative % variance	36.9	66.5	82.3	

Table 4 shows the factor loadings from factor analysis for three components accounting for 82.3% of the total variance for $PM_{2.5}$. The first component accounted for 36.9% of the variance and had high loadings on Cu, Zn, Sn, Pb (0.90–0.78) and moderate loading on Sb (0.59). These elements are typically tracers of industrial emissions. However, these elements have also been reported from vehicular emissions (Lin et al., 2005 and references there in). Zn is emitted from lubricant oil, brake linings, and tires (Adachi and Tainosho, 2004; Blok, 2005; Hjortenkrans et al., 2007), hence road traffic was found to be a key contributor to Zn particles (Lin et al., 2005). Average Cu/Zn ratios measured in all the sites was 0.09±0.13 suggesting vehicular sources (Cadle et al., 1999). It has been reported that Cu and Sb are used for break wear (Weckwerth, 2001; Salma and Maenhaut, 2006) and Sn is present in alloys used for automobile parts prone to wear such as automobile bearings. Strong apportioning of Sn to this factor suggests that vehicle abrasion is also responsible of emissions of this metal into the atmosphere. Sternbeck et al. (2002) proposed diagnostic criteria of concentration ratios between Cu and Sb (4.6±2.3) for brake wear particles. Table 5 compares Cu/Sb ratios reported in the literature with this study. The difference in the brake pad composition and type of vehicle (heavy duty or light duty) can be used to explain the variations in the ratios. Hence, it is concluded that this factor is associated with vehicular road dust originating from abrasion of vehicular parts and industrial emissions. The second factor explaining 29.6% of the variation and lists the same set of elements as present in factor 1 for PM_{10}. The correlation pattern for Ni, Cr, Sb and Mo (see the SM, Table S3) is different from the pattern for PM_{10} and only Ni and Cr (0.90) indicate a high degree of correlation. Sb–Cr (0.52), Sb–Ni (0.52), Sb–Mo (0.67) and Cr–Mo (0.70) show moderate degrees of correlation. This factor had high loadings for Cr, Ni, Mo (0.99–0.71) and moderate loading for Sb (0.54). This factor is apportioned to exhaust gases of cars (petroleum, diesel combustion) and municipal waste incineration, partly similar to the first factor of PM_{10}. The third factor explains 15.8% of the variation and had high loading of Cd (0.98) and Pb (0.61). This factor is attributed to coal combustion and non–ferrous metal production.

***Table 5.** Cu/Sb ratios to trace the road traffic source*

Material	Cu/Sb
Road traffic dust [a]	4.6±2.3
$PM_{2.5}$ from urban traffic tunnel [b]	7.0±1.3
Urban PM [c]	8.0±1.5
Road dust [c]	6.8±1.9
Break lining and rubber from tyre [d]	5.3
Automotive brake abrasion dusts [e]	12.7
Road dust (coarse PM) [f]	9.1±1.8
Road dust (fine PM) [f]	2.17±0.83
Urban $PM_{2.5}$ (this study)	8±1.2

[a] Sternbeck et al., 2002; [b] Fabretti et al., 2009; [c] Amato et al., 2011; [d] Hjortenkrans et al., 2007; [e] Iijima et al., 2007; [f] Lin et al., 2005

It is interesting to note that Pb is apportioned to two factors in both PM_{10} and $PM_{2.5}$ fractions indicating two distinct sources of the element in the atmosphere. We rule out petroleum as a major source of lead in atmosphere as leaded gasoline was phased–out in India from the beginning of this century. While one source of Pb could be coal combustion from hundreds of brick kilns along Hoogli River, the thermal power plants, cement factories etc, the other source can be the smelters from the medium and small scale industries around the city. Sb is strongly apportioned to one factor in the PM_{10} fraction but have two sources in the $PM_{2.5}$ fraction. While one source of Sb in the urban atmosphere can be brake wear as observed in numerous previous studies (Weckwerth, 2001; Salma and Maenhaut, 2006; Tian et al., 2012), the other source could be exhaust gases of cars and fuel oils (Tian et al., 2012), and municipal solid waste incineration (Watanabe et al., 1999; Tian et al., 2012).

3.5. Risk assessment from carcinogenic metals

Among the particle bound heavy metals in the Kolkata metropolitan area, chromium (VI), nickel, cadmium and lead are the known carcinogenic metals that are introduced by exposure through the inhalation pathway (Hieu and Lee, 2010).

Cr(VI) is classified as Group A, a confirmed human carcinogen by inhalation route of exposure (U.S. EPA, 2005). In this study the measured total Cr concentration was assumed to be a mixture of carcinogenic Cr(VI) and non–carcinogenic Cr(III) in 1:6 concentration ratio in ambient air (U.S. EPA, 2004). Hence for the carcinogenic risk assessment, we used a concentration of Cr(VI) as one seventh of the total concentration of measured Cr (U.S. EPA, 2004; Park et al., 2008; Hieu and Lee, 2010; Massey et al., 2013). Nickel refinery dust and nickel subsulfide (Ni_3S_2) are classified as Group A material, known human carcinogens by U.S. EPA. Cadmium is classified as Group B1, probable human carcinogen based on limited evidence from occupational epidemiologic studies. Pb is classified as B2, a probable human carcinogen but due to inadequate human evidence its unit risk is currently under amendment by U.S. EPA. Hence for this study the ECR of Pb was not calculated. The estimated ECR for the average value and 95th percentile value of $PM_{2.5}$ bound carcinogenic metals in the Kolkata metropolitan area is listed in Table 6. The ECR is multiplied by 10^6 for comparison with one in a million standards. Cr(VI) has the highest ECR. ECR posed by Cd and Ni are in the same range though the elemental concentration in $PM_{2.5}$ is quite different. The total ECR resulting from the exposure of the carcinogenic elements through inhalation pathways is 163 (average) and 361 (95th percentile). In other words, 163 people out of 1 million are at risk of developing cancer from exposure to the carcinogenic metals in the $PM_{2.5}$ fraction of the air particulates in the Kolkata metropolitan area.

Table 6. *Excess cancer risk of carcinogenic elements in $PM_{2.5}$*

Metal	Average (ng/m^3)	95th Percentile (ng/m^3)	Inhalation Unit Risk ($\mu g/m^3$)	Excess Cancer Risk (in 1 million)	
				Average	95th percentile
Cr(VI)	12	26.6	$1.2x10^{-2}$	144	319
Ni	40	77.4	$2.4x10^{-4}$	10	19
Cd	5	12.8	$1.8x10^{-3}$	9	23
Total				163	361

4. Conclusions

Among the Asian countries, India is the third largest economy after China and Japan. Extremely high rate of urbanization and industrialization in the country has very often crossed the fine boundary between development and sustainable growth. Many of the Indian metropolises are marred by air pollution including the national capital Delhi. Kolkata is no exception. Extremely high PM concentrations (PM_{10}=445 $\mu g/m^3$ and $PM_{2.5}$=313 $\mu g/m^3$) are observed during the winter months in Greater Kolkata, the financial hub of Eastern India. Elemental concentrations of the PM show anthropogenic sources for Ni, Cr, Cu, Sn, Zn, Mo, Sb, Cd and Pb based on EF calculations. The toxic and carcinogenic heavy metals (Cr, Ni, Zn, Mo, Sn, Sb, V, Co, Cu, Cd and Pb) are concentrated more in the breathable $PM_{2.5}$ fraction. Factor analysis performed on 9 elements with EF>10 apportioned the following sources for the PM: municipal waste incineration, vehicular emissions (exhaust gases, abrasion of automobile parts), industrial emissions, coal combustion and non–ferrous metal smelting. Carcinogenic risk assessment related to the respirable fraction of the PM was evaluated on the basis of metal concentrations in the $PM_{2.5}$. Chromium was found to be most risky in carcinogenicity among the $PM_{2.5}$ bound metals. Monitoring the concentration of toxic metals in the respirable size fraction ($PM_{2.5}$) of the PM is of paramount importance as the metals are transported into the lungs and eventually into the blood stream. This study provides a quantitative understanding of the metal concentration in the breathable (PM_{10}) and respirable ($PM_{2.5}$) size fraction of PM of a heavily polluted Indian metropolis and their possible sources. In many western countries mitigation approaches like sweeping, water flushing and chemical suppressants are used to prevent resuspension of vehicular emissions related road dust (Amato et al., 2010), however the practical aspects and effectiveness of using such methods in India are yet to be tested.

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Supporting Material Available

Chemical Analysis (Text S1), Description of sampling locations with GPS coordinates (Table S1), Correlation coefficients (*r*) among trace elements in PM_{10} and $PM_{2.5}$ (Table S2 and Table S3). This information is available free of charge via the internet at http://www.atmospolres.com

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