Identification of Sources of $PM_{2.5}$ at Farmgate Area, Dhaka Using Reconstructed Mass Calculation and Statistical Approaches

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Abstract

A study on air pollution at Farmgate, one busiest place of Dhaka city, Bangladesh was carried out by measuring fine air particulate matter of diameter 2.5 µm ($PM_{2.5}$) and determining its elemental and black carbon (BC) concentrations. $PM_{2.5}$ samples collected from the site during the period, from December, 2012 to February, 2013 were gravimetrically measured and the elemental and BC analyses of these samples ($PM_{2.5}$) were performed using PIXE/PIGE and Reflectance methods, respectively. The experimental data of elements BC and $PM_{2.5}$ masses were used to reconstruct the particulate mass by estimation of sources such as soil, smoke sea salt, sulfate and organic content. The sources of elements and BC in $PM_{2.5}$ samples were identified following RCM. From enrichment factor calculation, anthropogenic contribution of different elements (i.e., Na, Mg, Al, Si, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br and Pb) has been identified. The EF values of Mn, V and Ni are found to be 5.09, 7.33 and 9.16 respectively indicating that there were some anthropogenic contribution. Pearson's correlation matrix was generated in order to identify the rotations among the sources of fine air particulate matter. This study revealed that toxic heavy metals (i.e., Cr, Mn, Fe, Co, Ni, Cu, Zn and Pb) concentrations in the samples were varied significantly (%RSD: 44% to 145%) and presented non-homogeneously in different sampling times, which suggesting that these heavy metals, i.e., Cr, Mn, Fe, Co, Ni, Cu, Zn and Pb might come in the samples due to anthropogenic activities. Subsequently, principal component analysis was used to identify major elements associated with sources. Based on overall findings, it has been suggested that the anthropogenic sources (i.e., smoke, motor vehicles, petro-chemicals, brick industries, sea salt spray and soil dust) could be contributing contaminants in atmospheric particulate matter ($PM_{2.5}$) samples in the study area.

Keywords**:** PIXE, PIGE, $PM_{2.5}$, BC, RCM and PCA

1. Introduction

Urban air pollution a growing problem has become one of the most serious concerns especially vie of adverse health effects that has been associated with ambient fine air particulates [1-6]. In May 2016, the World Health Organization (WHO) reported that more than 80% of the urban areas had air pollution level higher than the recommended limit for health reasons particularly in low and middle income countries where it pollution increased upto 98% [7]. However, the contribution of anthropogenic sources to aerosol loadings has increased rapidly, especially over urban and industrial cities due to increasing industrial development and urbanization in Bangladesh [8].

Asian dust particles are transported over long distances by westerly winds in the late winter and spring seasons, substantially enhancing the ambient aerosol concentrations in downwind regions and countries. Along with windblown dust, tremendous amounts of pollutants accompanying these dust-rich atmospheric systems are delivered out of the Indian sub-continent, primarily because of increased development in industries and household coal combustion [9-10].

Fine particles that are very small and light tend to stay longer in the air than the heavier ones and naturally increased the chances of inhaling them into the bodies by humans and animals. Owing to their minute size, particles smaller than 2.5 micrometers are able to bypass the nose and throat and penetrate deep into the lungs and some may even enter the circulatory systems. Studies have found a close link between exposure to fine particles and premature death from heart and lung disease. Fine particles are also known to trigger or worsen chronic disease such as asthma, heart attack, bronchitis and other respiratory problems. The daily average of $PM_{2.5}$ level usually exceeded the Bangladesh national ambient air quality standard (NAAQS) of 65 µg/m^3 in winter in Dhaka, Bangladesh [11].

Urban air pollutants arise from a wide variety of sources including mainly combustion processes, emissions from motor vehicles and biomass burning. Epidemiological links have been established between fine airborne particles (< 2.5 µm in diameter) and the morbidness and mortality rates in large population [12] at airborne particle concentrations in the range of 10 to 30 µg/m^3. Different types of non-destructive nuclear techniques like, accelerator-based ion beam analysis (IBA) techniques are found well-suited to non-destructive, multi-elemental analysis of air filters in ambient aerosol studies [13-15]. PIXE and PIGE in aerosol studies have received great interest globally in conjunction with other nuclear methods commonly known as ion beam analysis (IBA) technique that are well-suited to particle and aerosol filter analysis [13, 16].

One of the simplest ways to carry out source apportionment analysis is reconstructed mass calculation method [17] a multivariate statistical technique called principal component analysis (PCA) is used in environmental studies to identify sources from data taken at receptor sites [18-20]. Principal component analysis generates the underlying "component" that describes groups of variables. In

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environmental studies, each component is generally treated as a source. The common version of principal component analysis (PCA) analysis uses the correlation matrix to calculate a set of factors that are rotated to generate a matrix of "factor loadings" for the variables.

To keep the environment clean and to protect the human from trace metal contamination, it is important to gain an understanding of the nature and the extent of trace metal pollution caused by input of dust. On the other hand, the identification of various sources of airborne particulate matter (PM) and the assessment of their impact on the aerosol composition of an air shed are some of the major goals of contemporary atmospheric research [21]. The measurement of the elemental concentration of atmospheric particles are needed for monitoring and studies of environmental deposition, and source attribution by reconstructed mass calculation and statistical analysis. Although there are few studies of heavy-metal contamination in urban dust in Bangladesh, but a little information is available on the extent of heavy metal contamination, its temporal variation, and the source of pollutants in particular matter (PM) samples.

However, the specific objectives of this study were (1) determine the concentrations of elements (As, Cd, Cr, Co, Cu, Mo, Ni, Pb, S, Zn, and Zr), and black carbon (BC) concentration in airborne particulate matter ($PM_{2.5}$) samples collected from one of the busiest places in the capital city of Bangladesh (Farmgate, Dhaka), (2) assessment of source attribution by reconstructed mass calculation using some elemental composition of atmospheric particles in the airborne particulate matter ($PM_{2.5}$) samples, and (3) identify sources of pollutants in the airborne particulate matter ($PM_{2.5}$) samples

2. Materials and Method

2.1 Site and sample collection

Farmgate (latitude: 23.76º N; Longitude: 90.39º E), one busiest place of Dhaka city, Bangladesh (Fig. 1) was selected for PM sampling due to its proximity from the intersection of several road-ways and a huge number of vehicles playing through the place, the area is a mixed zone of various commercial and semi-industrial establishments and academic institutions (Schools, Colleges and Universities). Besides, the Tejgaon industrial area is very near the area. For sample collection spot/station was taken on the roof of the guardhouse of Bangladesh Agriculture Research Council (BARC) within the study area. It's needed to be mentioned that this location/spot has been used as a continuous air monitoring station, known briefly as CAMS by the Department of Environment (DOE), Dhaka, Bangladesh since 2009, A portable Air Metrics MiniVol Sampler (Airmetrics, USA) was placed on the roof of the guardhouse of BARC for collection of $PM_{2.5}$ sample and the sampling was performed four times a week during the period from December, 2012 to February, 2013. The miniVols were programmed to sample at 5 lpm (litre per minute) through $PM_{2.5}$ particle size separator (impactor) and then through 2 µm pore teflon and quartz filters in two samplers. The actual flow rate should be 5 lpm at ambient conditions for proper size fraction. To ensure a constant flow of 5 lpm through the size separator at different air temperatures and ambient pressures, the sampler flow rates were adjusted for the ambient conditions at the sampling site. The MiniVol sampler was positioned with the intake upward and located in an unobstructed area at least 30 cm from any obstacle to air flow and the sampler inlet was placed at a height of 10 m above ground level. The intake manifold at the Farmgate location was located about 5 m away from the main road. Two samples of $PM_{2.5}$ were simultaneously collected on teflon and on quartz filters for 24 h with two MiniVol samplers.

2.2 Meteorological condition

Bangladesh has a climate of tropical monsoon, mild winter (October to March), hot, humid summer (March to June) and humid warm rainy monsoon (June to October). January is the coolest month with temperature averaging 26℃ and April is the warmest month with temperature ranging from 33 to 36℃. It rains mostly during June to October. From November to February, the general directions of the winds are north-easterly in the northern region, north-westerly over the rest of western Bangladesh, and northerly in the eastern part. From March to May, the winds are westerly to south-westerly in the western half but south-south, south-easterly in the eastern half. From June to September, the winds are not all southerly; much of it is either south-easterly or easterly [22].

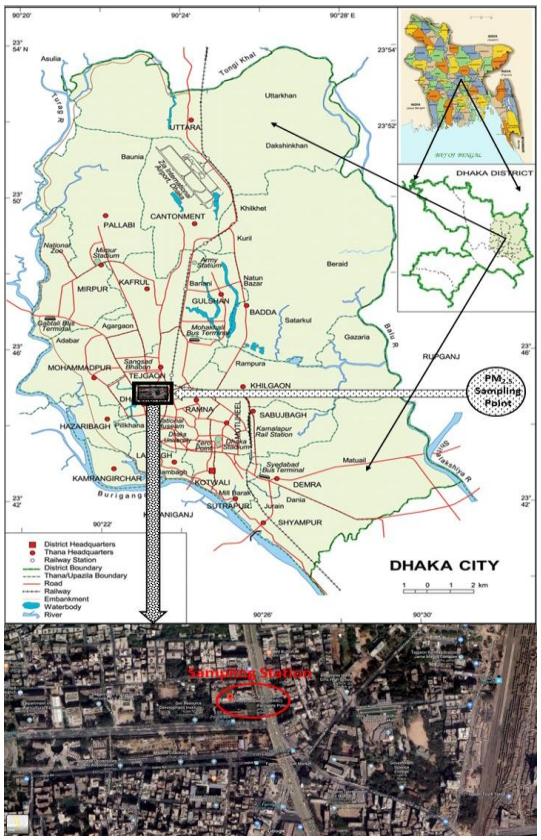

Fig. 1: Particulate matter ($PM_{2.5}$) sampling site: Farmgate , Dhaka, Bangladesh (Latitude: 23.76°N; Longitude: 90.39°E)

2.3 $PM_{2.5}$ mass and black carbon (BC) measurement

Particulate matter (PM) mass was estimated in the Atmospheric and Environmental Chemistry Laboratory of Atomic Energy Center, Dhaka (AECD), Bangladesh. $PM_{2.5}$ was determined by weighing the filters before and after exposure utilizing a microbalance (MT 5 Metler, Japan) maintaining room temperature approximately at 22°C and relative humidity at 50%. All filters were equilibrated at constant humidity and temperature of the balance room before and after every weighing. A U-shaped electrostatic charge eliminator (Mettler Toledo™, Japan) was utilized to remove the static charge gathered on the channels previously. The contrast in weights for each channel was ascertained and the mass focuses for each $PM_{2.5}$ tests were resolved. The $PM_{2.5}$ samples collected on teflon filters were analyzed by an EEL-type smoke stain reflectometer (Diffusion Systems Ltd., UK) to measure black carbon (BC) in $PM_{2.5}$ samples. Secondary standards of black carbon were used to calibrate the reflectometer [23]. The concentrations are defined based on the amount of reflected light absorbed by the filter samples and an assumed mass absorption coefficient. The concentration of BC is related to the concentration of light absorbing carbon. Iron (Fe) has a moderate light absorption coefficient and can have some limited influence on the BC value measured by reflectance. The uncertainty associated with the BC measurement is rather high (4% to 9%), and therefore, the influence of variation in Fe concentration on BC measurement is negligible [11].

2.4 Multi-elemental analysis

Multi-element analysis of the air particulate samples was made using Ion Beam Analysis (IBA) method at the Institute of Geological and Nuclear Science (IGNS), New Zealand. Data from these methods were collected simultaneously using a 2.6-MeV proton beam from a Van de Graaff accelerator. Beam diameters were typically 8 mm, and currents were kept at less than 2 nA/mm^2 to avoid beam damage to the filters and to reduce elemental losses. Run lengths were typically sufficient to deliver 30 μC (100 nA for 300 s) of total charge through the filter. Three nuclear analytical methods: (i) Proton-Induced X-ray Emission (PIXE), (ii) Proton-Induced Gamma Emission (PIGE), and (iii) Proton Elastic Scattering Analysis (PESA) are usually used for multi-elemental analysis [13, 24] in PM samples. However, for this study PIXE was used to analyze for 20 elements: Na, Mg, Al, Si, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, and Pb. On the other hand, PESA was used to analyze hydrogen (H) content in $PM_{2.5}$.

2.5 Reconstructed mass (RCM)

The reproduced mass or RCM is figured by expecting that six composite factors or pseudo sources, as given in the accompanying condition, are the real supporters of fine and coarse molecule mass [25].

RCM = [Soil] + [OMH] + [BC] + [Smoke] + [sulfate] + [Sea salt] + [Cu] + [Zn] (1)

Where the pseudo-sources are figured utilizing the basic convergences of their constituent components as given here:

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$$[Soil] = 2.20*[Al] + 2.49*[Si] + 1.63*[Ca] + 2.42*[Fe] + 1.94*[Ti] \quad (2)$$

$$[BC] = [Soot] \quad (3)$$

$$[Smoke] = [K] - 0.6*[Fe] \quad (4)$$

$$[Sea\ Salt] = 2.54*[Na] = [Na] + [Cl] \quad (5)$$

$$[Sulfate] = 4.125*[S] \quad (6)$$

The [Soil] factor contains components overwhelmingly found in the earth's covering (Al, Si, Ca, Fe, Ti) as oxides and incorporates a multiplier to remedy for the oxygen content and an extra multiplier of 1.16 to revise for the way that three real oxide patrons (MgO, K_2O, Na_2O), carbonate furthermore, bound water are rejected from Eq. (2).

On account of organic carbon [OC] assurance, add up to hydrogen on the channel is expected to include chiefly of H from natural material and ammonium sulfate. H is converted to OMH (organic carbon containing hydrogen) using following equation:

$$[OMH] = 11*([H] - 0.25*[S]) \quad (7)$$

Potassium, occur naturally in sea spray in mass ratios of 0.036 [26]. The ratio of chemical species present in sea spray can be used to define the non-sea salt component of various elements in the standard way: non-sea salt potassium where:

$$\text{Potassium } [nssK] = [Ktot] - 0.036 \times [Na] \quad (8)$$

2.5 Principal component analysis (PCA)

Principal component analysis (PCA) reduces the dimensionality of data by a linear combination of original data to generate new latent variables, which are orthogonal and uncorrelated to each other. It extracts the eigenvalues and eigenvectors from the covariance matrix of original variables [27]. PCA provides an objective way of finding indices of this type so that the variation in the data can be accounted for as concisely as possible [28]. PCA is often useful for providing information regarding source characteristics in terms of the elements that are associated with a given source type. These methods are based on the analysis of the correlation between measured concentrations of chemical species, assuming that highly correlated compounds are emitted from the same source. The experimental elemental data were subjected to analysis using IBM SPSS software (version 20).

3. Results and Discussion

Air quality (IAQ) has been a matter of public concern in Bangladesh especially for children and female as well as they are more sensitive to contaminants in environment. Therefore, different types of contamination in air: $PM_{2.5}$,

black carbon (BC), elements (i.e., H, Si, P, S, Cl, Br, Na, K, Ca, Mg, Al, Sc, Ti and V) and toxic heavy metals (i.e., Cr, Mn, Fe, Co, Ni, Cu, Zn and Pb) concentration in 45 samples collected from the one sampling point but different times around the year were analyzed by Proton-Induced X-ray Emission (PIXE). Subsequently, $PM_{2.5}$ was determined by weighing the filters before and after exposure utilizing a microbalance (MT 5 Metler, Japan) and the $PM_{2.5}$ samples collected on teflon filters were analyzed by an EEL-type smoke stain reflectometer (Diffusion Systems Ltd., UK) to measure black carbon (BC). However, specific discussion on particulate matter ($PM_{2.5}$), particulate mass: black carbon (BC) and elemental contents of $PM_{2.5}$ were made separately and described below:

3.1 Particulate matter mass ($PM_{2.5}$) and black carbon (BC) concentrations

Particulate mass was determined from the differences of unloaded and loaded filters. The 24 hours average mass values of $PM_{2.5}$ and their ratios with BC concentration from December 01 to 31, 2012 and 02 January to 27 February, 2013 obtained at Farmgate, Dhaka, Bangladesh were calculated, and the summary of the fine particulate matter mass and black carbon concentration for the study period mentioned above are given in Table 1.

Table 1: Summary of the fine particulate matter mass and BC concentration obtained at Farmgate site, Dhaka for the study period of December 2012 to February 2013

Sl No.	Parameter	$PM_{2.5}$	BC	BC/$PM_{2.5}$
1	Mean (µg/m^3)	159.07	32.28	0.203
2	Stdev (µg/m^3)	62.65	6.82	0.109
3	Maximum (µg/m^3)	299.26	46.95	0.069
4	Minimum (µg/m^3)	52.31	20.51	0.898
5	25% Percentile	107.31	27.93	0.260
6	Median (µg/m^3)	163.75	31.1	0.190
7	75% Percentile	200.72	37.72	0.188
8	VC (, µg/m^3)	0.39	0.21	0.538
9	GM (Geometric mean, µg/m^3)	146.52	31.57	0.215
10	%RSD (Relative Standard Deviation)	38.94	20.9	0.537
11	Skewness	0.383	0.237	0.619
12	Kurtosis	-0.498	-0.524	1.052

According to the study revealed that the average concentration of $PM_{2.5}$ for the period of 24 hours was found to range from 52.31 to 299.26 µg/m^3 with an average value of 159.07 µg/m^3. It was observed that the highest $PM_{2.5}$ concentration was 299.26 µg/m^3 on January 2, 2013, and the lowest $PM_{2.5}$ concentration was 52.31 µg/m^3 on February 17, 2013. To show the variation, box and whisker plot for monthly fine mass ($PM_{2.5}$) and BC concentration at sampling site during the study period are given below (Figs. 2 and 3). A two ways ANOVA (analysis of variance) test revealed that $PM_{2.5}$ and BC concentration for the study period was significantly different at a 95% confidence level.

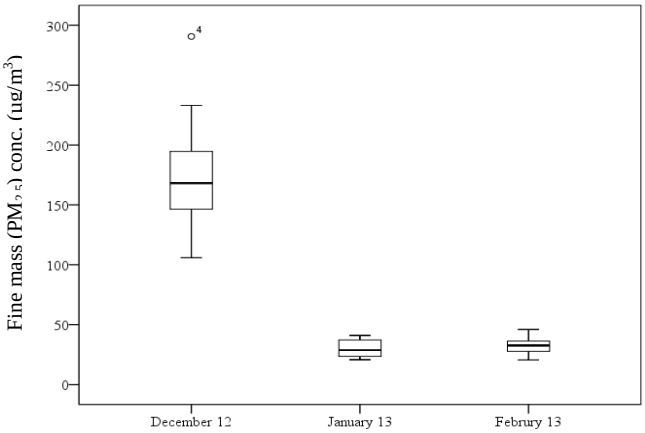


Fig. 2: Box and whisker plot for monthly fine mass ($PM_{2.5}$) concentration at Farmgate site during the study period

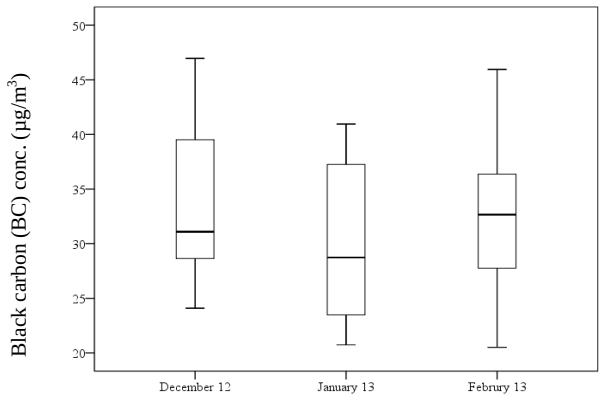
In this study, it was observed that the winter season's samples showed higher mass concentration. It might happen due to the reason that the mixing height became lower and the particulate matters were trapped nearer to ground level at the lower temperatures in winter season. During the winter, rainfall is minimal, therefore, dust resuspension increases the PM mass concentrations. Besides, the seasonal wind blowing from the north, and northwest may provide transported PM from India. On the other hand, air mass backward trajectories were calculated [29] using the vertical mixing model for those days indicated that higher contribution of concentration (both $PM_{2.5}$ and BC mass) for air particulate mass concentrations was also influenced by transboundary air pollution [30]. From this study, it was noticed that the values of the air quality index (AQI) in winter was very high and the air was classified as extremely unhealthy during this period [31].

Table 2: Variation of BC/ $PM_{2.5}$ ratio in different years from 1st to 14th January

Sl No.	Period (1st to 14th January)	BC/ $PM_{2.5}$ (with STD)
01.	2001	0.55 ± 0.15
02.	2002	0.43 ± 0.10
03.	2003	0.38 ± 0.04
04.	2004	0.36 ± 0.18
05.	2005	0.27 ± 0.05
06.	2006	0.39 ± 0.10
07.	2010	0.16 ± 0.03
08.	2013	0.14 ± 0.02

3.2 Reconstructed mass (RCM) variables

The sum of all the composite variables discussed in the previous section should provide a reasonable estimate of the total fine mass for comparison with the measured gravimetric mass of both fine particles on the filters. So, the



Sampling period: December, 2012 to February, 2013

Fig. 3: Box and whisker plot for monthly BC concentration at Farmgate site during the study period

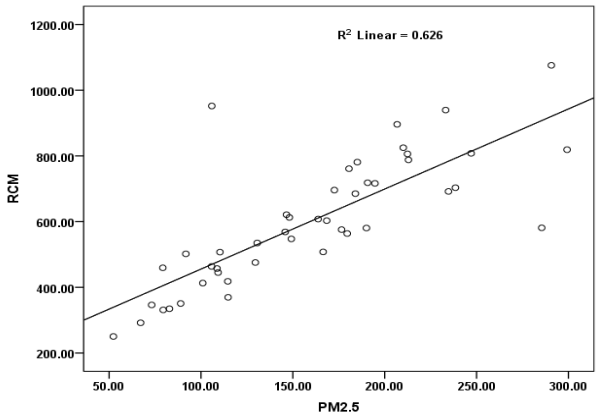


Fig. 4: The plot of RCM versus fine gravimetric mass during the sampling period

reconstructed mass (RCM) will be the sum of the reconstructed mass variables defined above. The RCM estimates here will be well below 100%, but still sufficient to do reasonable source apportionment estimates. It should be mentioned here that the number of elements measured spans the full range of possible chemical species found in most particulate matter collected here, as well as aluminum (Al), silicon (Si), calcium (Ca), iron (Fe), titanium (Ti), copper (Cu), zinc (Zn), potassium (K), sodium (Na), chloride (Cl), sulfur (S), soot are mainly present in particulate matter. The RCM was compared with the gravimetric weight of the filters, where the least squares fit to the data gave PCM = 0.54 × weight with an r = 0.626 in the case of fine mass. The reconstructed mass was about 50% in this study because nitrate and organic matter were not included in the calculation. However, this study revealed (Fig. 4) that the correlation between reconstructed mass the data gave PCM = 0.54 × weight with an r = 0.626 in the case of fine mass. The reconstructed mass was about 50% in this study because nitrate (RCM) and fine gravimetric mass during the sampling period was medium (r = 0.625).

3.3 Concentration of metals in $PM_{2.5}$ sample

The concentration of elemental (i.e., Na, Mg, Al, Si, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, and Pb) in 45 samples of $PM_{2.5}$ were analyzed by Proton-Induced X-ray Emission (PIXE) at the Institute of Geological and Nuclear Science (IGNS), New Zealand; and the statistical analysis for the elemental concentration in $PM_{2.5}$ samples is presented in Table 3.

Table 3: The mean and standard deviations (elemental concentration in µg/m^3) of fine particulate matter of Farmgate area, Dhaka, Bangladesh

Sl No	Element	Min	Max	Average	Stdev	%RSD	China[a]	Pakistan[b]	Hungary[c]	USA[d]	Japan[e]
1	H	15.27	67.42	36.09	13.16	36.45	--	--	--	--	
2	Si	5.27	25.69	14.57	5.17	35.52	--	--	--	--	--
3	P	0.60	1.88	1.14	0.30	26.06	--	--	--	--	--
4	S	23.66	73.12	48.49	13.47	27.77	--	--	--	--	--
5	Cl	1.58	74.41	21.74	17.38	79.96	--	--	--	--	--
6	Br	0.00	1.04	0.20	0.26	131.70	--	--	--	--	--
7	Na	0.00	39.85	10.09	10.95	108.52	1.60	1.90	0.45	--	--
8	K	5.90	33.10	16.90	5.97	35.31		5.60	0.42	--	--
9	Ca	0.95	6.91	2.69	1.26	46.74	9.05	9.10	2.60	--	--
10	Mg	0.00	1.76	0.71	0.45	63.28	2.04	2.10	0.48	--	--
11	Al	1.34	6.55	3.52	1.26	35.69	5.33	8.40	1.06	--	--
12	Sc	0.00	0.21	0.05	0.06	109.02	--	--	0.01	--	--
13	Ti	0.00	0.50	0.18	0.14	75.21	0.33	0.56	0.07	--	--
14	V	0.00	0.20	0.05	0.06	132.46		0.02	0.03	--	--
15	Cr	0.00	0.34	0.08	0.07	91.61	0.04	0.03	0.09	0.07	0.04
16	Mn	0.01	1.59	0.33	0.40	118.95	0.11	0.33	0.03	0.01	0.06
17	Fe	1.08	7.91	3.41	1.50	43.98	3.73	8.20	1.93	0.51	0.93

Sl No	Element	Min	Max	Average	Stdev	%RSD	China[a]	Pakistan[b]	Hungary[c]	USA[d]	Japan[e]
18	Co	0.00	0.21	0.03	0.04	130.85		0.03	0.04	--	--
19	Ni	0.00	0.20	0.05	0.06	118.17	0.04	0.02	0.03	0.02	0.01
20	Cu	0.00	0.44	0.10	0.10	97.14	0.05	0.07	0.06	0.01	--
21	Zn	0.62	40.20	6.01	8.73	145.39	0.33	1.10	0.08	0.02	0.54
22	Pb	0.00	25.72	5.41	6.61	122.25	0.11	4.40	0.02	0.01	0.21

[a][32], [b][33], [c][34], [d][35], [e][36]

Table 4: Pearson's correlation matrix among elements in the analyzed $PM_{2.5}$

Element	Na	Mg	Al	Si	P	S	Cl	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Br	Pb
Na	1.00																				
Mg	0.16	1.00																			
Al	0.23	**0.50**	1.00																		
Si	0.09	**0.36**	**0.81**	1.00																	
P	0.19	0.12	**0.66**	**0.80**	1.00																
S	0.27	-0.22	**0.17**	0.28	**0.66**	1.00															
Cl	0.18	-0.12	0.23	**0.35**	**0.62**	**0.71**	1.00														
K	0.17	-0.08	**0.39**	**0.42**	**0.74**	**0.72**	**0.66**	1.00													
Ca	0.23	0.23	**0.89**	**0.74**	**0.73**	0.35	**0.50**	0.54	1.00												
Sc	0.01	0.02	0.23	**0.48**	**0.32**	0.05	0.14	0.02	0.24	1.00											
Ti	0.29	0.29	**0.64**	**0.43**	**0.45**	0.29	**0.32**	0.23	**0.63**	0.08	1.00										
V	0.02	-0.15	0.21	0.13	0.27	0.22	0.12	**0.42**	0.25	-0.15	-0.05	1.00									
Cr	0.18	-0.19	-0.01	0.08	0.14	0.22	0.22	0.13	0.09	0.16	0.05	-0.22	1.00								
Mn	0.06	0.19	0.09	-0.04	0.18	**0.43**	**0.52**	**0.34**	0.16	-0.11	0.27	-0.11	0.02	1.00							
Fe	0.17	0.26	**0.74**	**0.66**	**0.69**	**0.38**	**0.68**	**0.56**	**0.87**	0.23	**0.62**	0.08	0.22	**0.34**	1.00						
Co	0.19	-0.03	0.04	0.15	0.00	-0.01	-0.11	0.07	-0.07	-0.08	0.20	0.20	0.16	-0.30	-0.14	1.00					
Ni	0.25	0.14	0.25	0.15	0.14	-0.02	0.14	0.21	**0.33**	0.04	0.18	0.11	0.19	0.00	**0.33**	-0.04	1.00				
Cu	0.03	0.05	0.26	0.21	0.26	0.13	**0.39**	0.28	**0.33**	0.05	0.08	0.23	-0.07	0.24	**0.48**	-0.17	0.02	1.00			
Zn	0.09	0.00	0.15	0.14	**0.39**	**0.51**	**0.60**	**0.46**	0.26	0.13	**0.32**	0.00	0.15	**0.65**	**0.41**	-0.21	0.11	0.04	1.00		
Br	-0.03	0.24	0.23	0.17	0.27	0.26	**0.41**	0.26	0.29	-0.02	0.23	-0.12	-0.03	**0.44**	**0.33**	-0.28	0.24	0.29	0.29	1.00	
Pb	0.17	0.18	**0.46**	**0.54**	**0.64**	**0.46**	0.37	**0.64**	**0.48**	0.25	0.28	**0.41**	-0.18	0.08	**0.42**	0.01	0.21	0.07	0.36	0.05	1.00

Bold digits are statistically significant for correlation at 95% confidence level

In this investigation, the concentration of toxic heavy metals (i.e., Cr, Mn, Fe, Co, Ni, Cu, Zn and Pb) in $PM_{2.5}$ samples were varied significantly (%RSD: 44% to 145%), and presented non-homogenity in different sampling times (Table 3), with the indication that these heavy metals, i.e., Cr, Mn, Fe, Co, Ni, Cu, Zn and Pb might be come in $PM_{2.5}$ samples due to anthropogenic activities [37-38]. It might be happened due to the reason that Cu, Zn, Pb, Fe are usually emitted in the environment through industrial activities and motor vehicles, which might be sources of Cu, Zn, Pb, and Fe in dust samples [39-45]. Similar trend was also found for the trace elements (i.e., Na, Mg, Al, Si, S, Cl, K, Ca, Sc, Ti, V and Br) determined for this study. Anyway, the average heavy metal concentrations for the studied metals in 36 indoor dust samples were compared with the reported global results, and the recommended data published in literature (Table 3). It was observed from this study that the heavy metals concentration in indoor dust samples were significantly different from one country/city to another country/city, and it might be happened due to impact of on-going activities around the major highways and due to different geo-chemical composition of soil in different countries [42].

3.4 Pearson correlation matrix

Pearson's correlation matrix was generated in order to identify the rotations among the sources of fine air particulate matter. The Pearson correlation matrix of different elements found in finer air particulate matrix is given in the Table 4.

Correlation matrix showed that inter-element relationships agree with the results obtained from PCA. It also showed some new associations among different elements that were not adequately reported in the previous sections. The *p* value of correlation matrix indicated the strength of association among different elements; such as, *p* value containing 0.01 and 0.05 indicated strong and significant correlations respectively. This study revealed that correlation between Na and Cl (r = 0.181) was very weak (Fig. 5). This weak

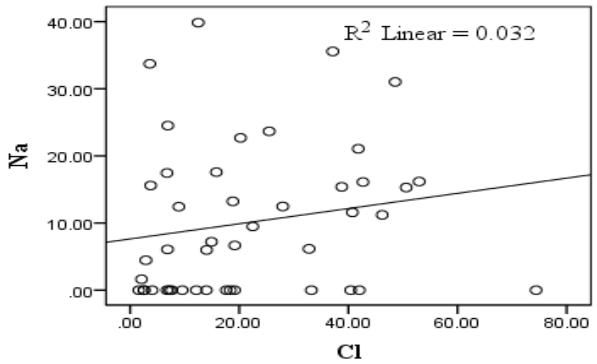


Fig. 5: Correlation plot for Cl vs. Na at Farmgate site for fine fraction

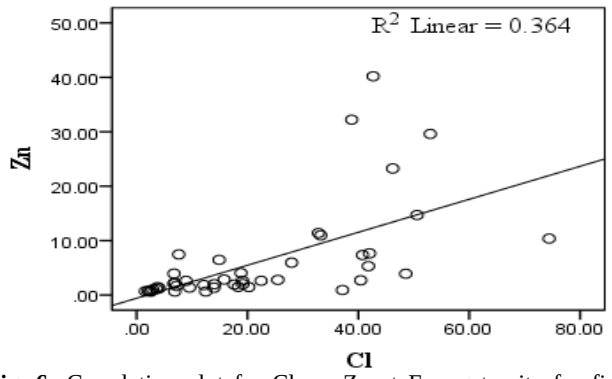


Fig. 6: Correlation plot for Cl vs. Zn at Farmgate site for fine fraction

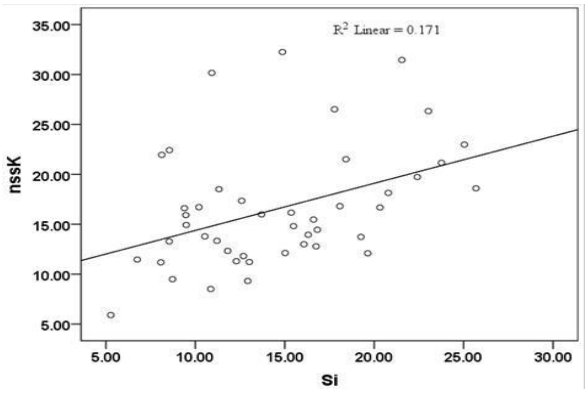


Fig. 7: Correlation plot for nssK vs. Si at Farm-gate site for fine fraction

association indicates that there was influence of anthropogenic sources except sea salt spray. Cl possessed significant association with S (r = 0.710), P (r = 0.615) and K (r = 0.663), this result also revealed that Cl had another anthropogenic sources except sea salt spray. The strong positive correlation (r = 0.737) of K with P and K with S (r = 0.717) could indicate similar source origin probably from biomass or waste burning emissions. Al displayed strong positive correlation with Si (r = 0.811), P (r = 0.658), Ca (r = 0.894), Ti (r = 0.635), Fe (r = 0.742) and significant correlation with Pb (r = 0.460). Si also showed strong correlation with P (r = 0.799), Ca (r = 0.737), Fe (r = 0.664) and significant correlation with Pb (r = 0.542), Sc (r = 0.477), Ti (r = 0.433). This observation suggested that all of these associated elements originated from soil dust. Fe also showed strong correlation with Ca (r = 8.70), Ti (r = 0.617) these could be originated from building materials, road and traffic dust. Pb also exhibited positive correlation with S (r = 0.456), P (r = 0.640) and K (r = 0.643). This result indicated that Pb possessed another source origin probably from biomass burning and waste burning except soil dust. Zn showed strong positive correlation with S (r = 0.510), Cl (r = 0.603) and Mn (r = 0.648) (Fig. 5). Among this association Zn and S emitted from motor vehicles as the sulfur contents of and gasoline were 0.7% and 0.2% respectively. Zn could be emitted from galvanizing factory or from motor vehicles; the strong association of Zn and Mn and significant correlation of Zn and Fe (r = 0.407) revealed this fact. The strong correction between Zn and Cl (r =0.630) and significant correlation of Zn with K (r = 0.462) and P (r = 0. 390) indicated that Zn possessed contribution to the origin of smoke content. Br showed significant correlation with Cl (r = 0.410) indicating similar sources of origin probably from sea salt spray or municipal incineration. The major source of Na and Cl is sea salt spray [46] but the correlation plot (Fig. 5) showed that they had another source other than sea salt spray. This phenomenon was explained in both Pearson correlation matrix analysis and principal component analysis. Chloride (Cl) possessed correlation with Zn (Fig. 6). Another significant multi sources element, Potassium (K) has three major sources: Smoke from biomass burring, sea salt and wind blown soil [47]. The biplot of non sea salt potassium (nssK) with major crustal element (Si) (Fig. 7) and black carbon (BC) (Fig. 8) confirmed that K mainly originated from biomass burning with black carbon rather soil dust.

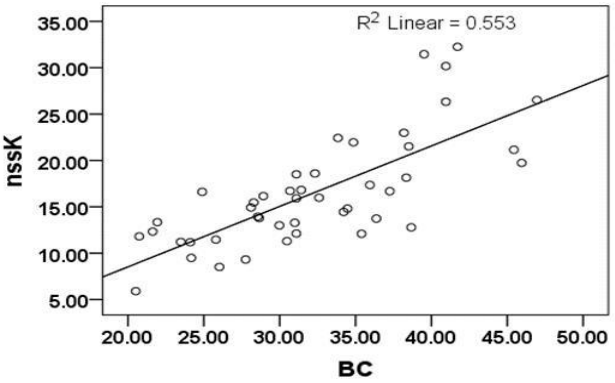


Fig. 8: Correlation plot for nssK vs. BC at Farmgate site for fine fraction

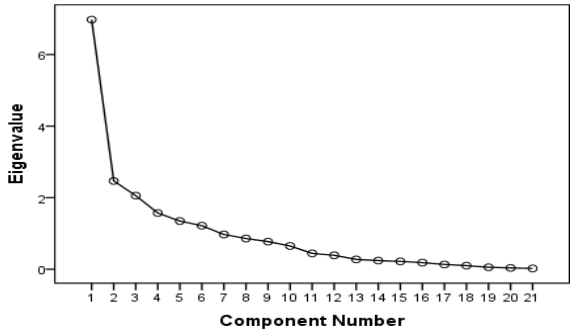


Fig. 9: Scree plot of the characteristic roots of principal component analysis

3.5 Enrichment factor (EF)

The enrichment factor (EFcrust) is used to define the origin of the metals in aerosol. EFc (enrichment factor from crust to air) = (Cx/CFe)air/ (Cx/CFe)crust. Here; (Cx/CFe) air: the concentration ratio of the element X and Fe in the measured air. (Cx/CFe) crust: the abundance of the element X and Fe in the crust. The elements have EFc values which range between 1 and 10, indicating that they are present in the aerosol in roughly crustal proportions. Usually, Na, K, Al, Mg, Ca, Mn and Fe are used as the reference metals. Fe was shown to be the best correlated with all other elements hence, selected as the reference element. The crustal abundance of elements was taken from list of abundance of chemical elements in earth's crust.

Except Ti, the other heavy metal concentrations were independent of Fe concentrations. So, it could be said that their source were non-crustal. Anthropogenic contribution on heavy metal concentrations were observed as Pb>Zn> Cu>Cr>Ni>Mn>V>Ti in a decreasing order respectively. The EFc values of Pb and Zn were found to be 5666.10 and 1174.97 respectively indicating the high anthropogenic contribution. Cu possessed EFc value of 29.32 indicating that the main source of Cu was anthropogenic. Having EFc value 11.87, Cr revealed that anthropogenic source could be considered along with natural occurrence. Subsequently this study narrated that the EFc values 5.09, 7.33 and 9.16 found for Mn, V and Ni respectively, which indicated that there were some anthropogenic contribution [9].

3.6 Principal component

In order to obtain reliable estimates of the different sources contributing to the fine mass measured at the Farmgate site, principal component analysis (PCA) was used to identify major elements associated with sources. Varimax rotation was used to maximize the sum of variances of the factor coefficients, which better explained the possible groups/factor that influenced fine air particulate matter. The total variance in each factor was calculated as the sum of the squared loadings for the given factor. A plot of this eigenvectors as a function of factor number is shown in the Fig. 9. There were six factors with eigenvalues greater than 1.0 for the fine particle data seta, which represented 74.46% of the total variance (Table 5). The scree plot was used to identify the number of PCs to be retained to understand the underlying element structure. The calculated factor loadings together with cumulative percentage and percentage of variance were explained by the each factor as listed in Table 5.

The positive and negative scores in PCA indicated that most of the air samples were either essentially affected or unaffected by the presence of extracted loads on specific factor/component, respectively. About 54.75% of the total variance was represented in the first three loadings factor. In this study, PC1, PC2, PC3, PC4, PC5 and PC6 explained more than 33, 11, 9, 7, 6, 5% of the total variance respectively (Table 5).

The first principal component (PC1) in the fine air particulate matter ($PM_{2.5}$) data sets explained more than 33.21% of the total variance. It was loaded with Si, Al, Ca, Fe, P, Ti, Sc, Mg and Pb. These elements are typical indicators of soil (Table 6). Among these element Pb is not actual earth crustal element. PCA analysis showed high factor loadings of Pb. Although leaded gasoline was banned in Bangladesh in 1999, Pb might be come from fugitive sources, for example paint and pigment, Pb acid battery [48] or the re-suspended soil dust [49]'

Table 5: Principal Component Analysis (PCA) with varimax rotation for all $PM_{2.5}$ data from Farmgate area, Dhaka, Bangladesh

Rotated Component Matrix						
Element	Component					
	PC1	PC2	PC3	PC4	PC5	PC6
Si	.908	.203	-.117	-.066	.062	.054
Al	.904	.069	.042	.233	-.137	.124
Ca	.810	.279	.083	.221	.015	.289
Fe	.712	.323	.287	.200	.170	.348
P	.683	.625	.002	-.009	.053	.106
Ti	.595	.114	.362	.300	.007	-.094
Sc	.520	.033	-.079	-.463	.431	-.144
Mg	.513	-.334	.340	.252	-.350	-.238
S	.096	.876	.162	.057	.119	-.005
K	.280	.830	.006	.146	-.074	.214
Cl	.214	.726	.347	.072	.235	.276
Zn	.119	.605	.550	.043	.062	-.237
Pb	.525	.589	-.122	-.032	-.341	-.149
Mn	-.057	.401	.772	.114	-.055	.007
Co	-.010	.062	-.638	.316	.031	-.139
Br	.163	.105	.626	.113	-.010	.323
Na	.144	.177	-.073	.686	.104	-.209
Ni	.223	-.008	.023	.576	.113	.184
Cr	-.006	.151	-.082	.274	.832	-.001
V	.047	.427	-.452	.092	-.510	.349
Cu	.189	.103	.191	-.037	-.052	.798
Eigen value	6.975	2.467	2.056	1.573	1.348	1.218
Variance %	33.215	11.749	9.792	7.488	6.420	5.799
Cumulative %	33.215	44.964	54.756	62.245	68.644	74.463

Table 6: Highly loaded elements (>0.5) in different principal component with their probable source**s**

Soil	Smoke	Motor vehicles	Sea salt	Cr-source	Cu-source
Si	S	Mn	Na	Cr	Cu
Al	K	Br	Ni		
Ca	Cl	Zn			
Fe	P				
P	Zn				
Ti	Pb				
Sc					
Mg					
Pb					

The second principal component analysis (PC2) explained more than 11.74% of the total variance. It was highly (> 0.5) loaded with P, S, K, Cl, Zn and Pb (Table 5). Among them K originated from biomass and wood burning (brick kiln). This component also had a high value of S that most likely originated from anthropogenic sources like energy production, biomass burning; refuse incinerations, emissions from ships using heavy oil. Again, sulfur can serve as a tracer for traffic exhaust in Bangladesh, as there is no coal- fired power plants in the country. Chlorine (Cl), phosphorus (P) and lead (Pb) were found to be major fine fraction components from pyrogenic emissions (Fig. 10). Except sea salt spray chlorine (Cl) might come mainly from municipal incineration.

Another high factor loading zinc (Zn), which might come as smoke from zinc chloride smoke mixture used as smoke grenade in military purpose. Generally all of these elements are exposed to the air like as exhausted smoke. So these elements are indicators of smoke. In the PCA results, it was not possible to separate this area sources from the smoke emissions.

PC3, accounting for more than 9.79% of total variance, was associated with Zn, Mn and Br indicating motor vehicles factors (Table 5). Zinc (Zn) is emitted from lubricating oil combustion in two-stroke engines [50]. Although three wheeled taxis with two stroke engines have been completely banned since January 2003, but unfortunately personal two-stroke motorcycles and scooters are still in use, leading to continued Zn emission [30]. Tire wear of motor vehicles are another source of Zn emission [51]. Combustion exhaust gases from motor vehicles contain manganese oxides (MnO_2). Bromine (Br) is used as fuel (gasoline, diesel) additives. So, Bromine (Br) also originated from motor vehicles exhausted (Table 6).

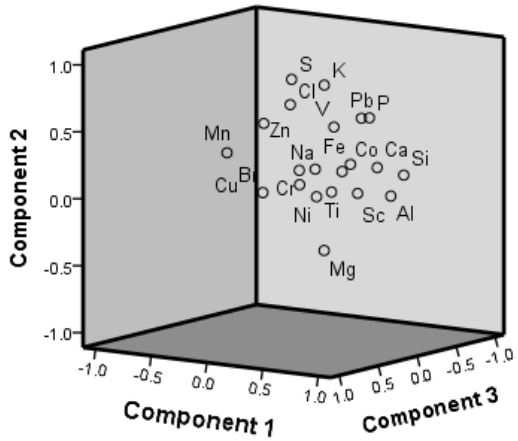


Fig. 10: Component plot in rotated space of principal component analysis

Principal component, PC4 explained more than 7.48% of total variance and highly loaded with Na and Ni (Table 5). Generally, sodium (Na) exposed to air as NaCl from sea salt spray. But in this component the loading of chloride (Cl) was very low. Generally chloride (Cl) could volatilize from aerosol or from filters in the presence of acidic aerosol, particularly in the fine fraction via the following reactions.

$$NaCl\ (s) + HNO_3\ (aq) \rightarrow NaNO_3\ (s) + HCl\ (g)$$

$$NaCl\ (s) + H_2SO_4\ (aq) \rightarrow Na_2SO_4\ (s) + HCl\ (g)$$

Another highly loaded element in this component was nickel (Ni). Nickel could be originated from sea salt spray. The average nickel (Ni) content of seawater is 1.7 micrograms per liter [52]. It is to be noted that the air particulate material was collected during the winter season but sea salt source generally is more dominated during the monsoon season when the wind is normally from the south and southeast (Table 6).

Principal component, PC5 and PC6 explained more than 6.42% and 5.79% of the total variances respectively (Table 5). These two components are labeled as chromium (Cr) source and copper (Cu) source respectively due to high loading of these two elements in their respective component (Table 6). Chromium (Cr) generally exposed to the air from different industries [53-54]. The metal chromium is used mainly for making steel and other alloys. Chromium compounds, in either the chromium (III) or chromium (VI) forms, are used for chrome plating, the manufacture of dyes and pigments, leather and wood preservation, and treatment of cooling tower water. Smaller amounts are used in drilling muds, textiles, and toner for copying machines. The sources of copper (Cu) in the atmosphere are Cu-containing fungicides, metal working factories, electroplating materials, iron and steel industry, wire burning and incineration [55]. The highly loaded elements (>0.5) in different principal component (PC) are listed in the Table 6.

4. Conclusion

Fine particulate matter ($PM_{2.5}$) air pollution studies at one of the busiest places (Farmgate) of Dhaka City Bangladesh showed the 24 hour average $PM_{2.5}$ mass level from 52.31 to 299.26 $\mu g/m^3$, which is comparison with the WHO'S air quality guideline value (25 $\mu g/m^3$ 24 hour mean) was found to be much higher. So the air quality at the study area was appeared to be highly deteriorated and detrimental to human health. The value of the ratio $BC/PM_{2.5}$ obtained from the present study was found to be the lowest one (0.14 ± 0.02). Compared with those of the previous years, this study indicating that black carbon (BC) concentration in $PM_{2.5}$ was reduced. The study also provided many other information regarding anthropogenic sources of different toxic heavy metals, which aspect would be much useful for further studies as well as taking measure for controlling air pollution. Thusly, territorial endeavors will be essential alongside neighborhood control activities to enhance the air quality.

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