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CHARACTERIZATION OF CHITTAGONG AEROSOL BY PCA MODELING

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ABSTRACT

Black carbon and other selected trace elements concentrations in aerosol samples collected at the Continuous Air Monitoring Station (CAMS) in Chittagong, the second largest city in Bangladesh, were investigated for possible source contributions. The particulate matter (PM) sampling was done from end of winter to middle of rainy season (February and July, 2007) using dichotomous sampler. The samples collected in two fractions of <2.5 μm (fine) and 2.5 to 10 μm (coarse) were analyzed for elemental concentrations by proton induced X-ray emission (PIXE), hydrogen by proton elastic scattering analysis (PESA), and black carbon by reflectance measurement. The elemental data sets together with black carbon were analyzed by principal component analysis method to identify the possible sources contributing to the mass concentration of coarse and fine particulate matter (FPM) fractions. The best solutions were found to be six and seven factors for coarse and fine fractions respectively, which could explain more than 90% of the variance in the data set. The sources were identified as biomass burning/brick kiln, soil dust, road dust, Zn source, Pb source, motor vehicle, CNG (compressed natural gas) vehicle and sea salt. It was found that in coarse fraction, the sea salt is mixed with Zn source and in fine fraction, the road dust factor is mixed with CNG vehicle source.

Key words: Continuous air monitoring station, PIXE, PESA, PM, CNG

INTRODUCTION

It is well recognized that air pollution has hazardous effects on human health causing respiratory diseases (Dockery and Pope 1994, Dockery *et al.* 1989). It is also harmful to plants and vegetation resulting in negative economic impact on the agricultural sector (Agrawal *et al.* 2003). Particulate deposition on vegetation results in the physical smothering of leaf surface of the plants. This causes increased susceptibility to disease, reduces light transmission, and decreases photosynthesis. Pollution can also be harmful to soil depending on the composition of particles.

The particulate matters in the atmosphere are subjected to world-wide distribution by air mass circulation and they are ultimately returned to the earth's surface by wet deposition through precipitation and by dry deposition through sedimentation, impaction and diffusion. After their deposition on earth's surface, they enter in the food chain at a

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rate determined by the deposition intensity, bio-geochemical processes and hydrodynamic conditions of a given ecosystem (Agrawal *et al.* 2003). These trace elements in the environment can be considered either as nutrients, or as toxic pollutants and also as a tracer for transfer mechanism. Therefore, for the global mass balance model of these elements, it is necessary to study the regional as well as global distribution of trace elements.

Particulate air pollution can be generated by natural and anthropogenic activities. Anthropogenic sources can be stationary and mobile. It has been estimated in many countries, traffic-related emissions constitute more than 50% of the total particulate air pollution (EPA 1996). Diesel engines power land and sea transport provides electrical power and are used for farming, construction, and industrial activities. However, diesel engines pollute the environment and concern is growing to abate and contain diesel pollution affecting public health in urban areas where population concentration tends to be high.

In general air quality problem is more severe in urban areas compared to rural ones since anthropogenic factors tend to dominate due to economic activities. Although depending on the meteorological condition especially wind direction and speed adjacent peri-urban and rural areas may also be affected by the urban air pollution. To address the increasing air pollution situation it is always necessary for the policy maker to know the major source of air pollution and their contribution to undertake cost effective pollution control measures.

Chittagong (latitude 22.22N, longitude 91.47E) is the largest port city of Bangladesh (Fig. 1) and is heavily trafficked predominantly with old and worn out commercial vehicles; especially the central city area covering about 10 km^2. The main road network in the city goes towards the port area and northward towards the industrial areas.

The geographical layout of the city, with dispersed industrial areas mostly at significant distances from the port area, results in a high number of road kilometers with high diesel traffic and their emissions. The large number of auto-rickshaws (three-wheel, two stroke engine taxis) represent another significant air pollution source in the city. A considerable fraction of them already run on CNG because of cost effectiveness of the fuel although this is not yet a regulatory requirement for Chittagong vehicles. Steel re-rolling mills and brick kilns have substantial emissions that affect their immediate surroundings as well as the overall air quality of the port city. The largely uncontrolled steel and re-rolling mills are located within commercial and residential areas, resulting in substantial particulate matter exposure to the residents of those areas. The cement factories in Chittagong that only perform cement-mixing operations, may also affect their neighborhoods to some extent. Brick kilns are large emitters, especially since there are so many of them, about 200, located in two clusters on the northern side of the city.

The objectives to characterize and identify possible sources contributing to the PM samples collected from Khulshi area of Chittagong city considered to be heavily polluted by traffic-related emissions and other anthropogenic sources. This paper presents PM and its elemental composition data as well as an estimate of the major sources using chemometric analysis methods.

A Continuous Air Monitoring Station (CAMS) is operated in Chittagong to measure criteria pollutants. The location of the CAMS is in the Chittagong Television Station Campus at Khulshi, which is on a hilltop about 2.5 km northwest of the downtown and about 100 meters above the surrounding area (Fig. 1). The location thus is unaffected by nearby air pollution sources, and it is representative of the air pollution concentrations of the city.

Fig. 1. Map of Bangladesh showing the location of Chittagong.

MATERIALS AND METHODS

One hundred and four pairs of $PM_{2.5-10}$ and $PM_{2.5}$ samples were collected on 37 mm diameter Teflon filters using a Thermo Andersen dichotomous sampler (Model no. 241) at the Chittagong CAMS by the Air Quality Management Project (AQMP) staffs. The

samples were collected from February to July, 2007. Appropriate QA/QC protocol was followed during sampling and mass measurements.

The samples were transferred immediately to the CAMS laboratory, Chittagong, for mass measurement. The aerosol masses of both the coarse and fine fractions were determined by weighing the filters before and after the exposure. A Po-210 (alpha emitter) electrostatic charge eliminator (STATICMASTER) was used to eliminate any charge accumulated on the filters before each weighing. Black carbon (BC) was measured at the AECD laboratory in Dhaka using an EEL (Evans Electroselenium Limited) Smoke Stain Reflectometer (Biswas *et al.* 2003). Secondary standards of known black carbon concentrations are used to calibrate the reflectometer. The concentrations are defined based on the amount of reflected light that is absorbed by the filter sample and an assumed mass absorption coefficient. It is related to the concentration of light absorbing carbon through standards of carbon with known areal density.

Multielemental analyses of the samples collected during the timeframe as mentioned above were made using proton induced X-ray emission (PIXE), Proton Induced Gamma Emission (PIGE) and proton elastic scattering analysis (PESA) techniques at GNS Science, Lower Hutt, New Zealand (Begum *et al.* 2004). X-ray spectra obtained from PIXE measurements were analyzed using the computer code GUPIX developed by Guelph University. Calibration of the PIXE system was performed by irradiating suitable micromater thin target elemental standards. Na was determined using PIGE and hydrogen was determined using PESA method. The concentrations of 16 elements, black carbon, and mass were obtained from these multiple analyses.

RECEPTOR MODELING

In order to investigate the variation on the concentration profiles of different trace metals, the enrichment factors (EF) compared to the crustal abundances were calculated. The crustal abundance data were taken from Mason (Mason 1966). Fe was chosen as the reference. The enrichment factors (EF) are given by

$$EF_i = \frac{C_i}{C_{Fe}} / \frac{A_i}{A_{Fe}}$$

where,

EF_i = Enrichment factor for the element, i.
C_i = Observed concentration of the element, i.
A_i = Crustal abundance of the element, i.

The subscript “Fe” refers to the relevant quantities for Fe.

The PIXE analysis of PM samples provided a sufficient number of elements to develop fingerprints related to a variety of PM sources. It is useful to combine some of

these elements and estimate the concentrations of compounds likely to represent most of the measured element such as estimating the amount of ammonium sulfate from the measured sulfur concentration. Other combinations of elements that represent interesting aerosol components can be estimated using pseudo-elements such as "soil". Sulfate can exist in the atmosphere as sulfuric acid producing acid rain or be partially neutralized to ammonium bi-sulfate or fully neutralized to ammonium sulfate (Malm *et al*. 1994). It is assumed that sulfur occurs in the atmosphere as fully neutralized ammonium sulfate. Hydrogen in the particles is present because of both the ammonium sulfate and from the organic matter in the particles. The organic matter mass can be estimated using the procedure given by (Malm *et al*. 1994) in which the hydrogens associated with ammonium sulfate are subtracted from the total hydrogen concentration

$$OMH=11(H-0.25*S)$$

The details for the calculation of RCM has been described elsewhere (Begum *et al*. 2009). The reconstructed mass (RCM) is the sum of the reconstructed mass variables defined above:

$$RCM = (NH_4)_2\,SO_4 + salt + soil + Smoke + OMH + BC + Zn + Pb$$

where OMH represents the term for organic matter. This equation includes contributions from most of the major components measured in both the coarse and fine mass. The RCM estimates here will be well below 100% but it is sufficient to do reasonable source apportionment estimates as the number of elements measured spans the full range of possible chemical species found in most particulate matter collected here.

The process of identification and apportionment of pollutants to their sources is an important step in air quality management. Principal component analysis (PCA) (Hopke 1985) uses measurements of pollutant concentrations at a sampling site to identify significantly correlated variables. This method extracts components explaining the majority of variance of the data matrix that is then qualitatively interpreted as possible sources (Hopke 1985, Hopke *et al*. 1976, Wolff *et al*. 1985). PCA is often useful to provide 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.

RESULTS AND DISCUSSION

A total of 104 samples collected at CAMS Chittagong city in coarse ($PM_{2.5-10}$) and fine ($PM_{2.5}$) fractions were analyzed in the present study. Table 1 represents the basic statistics of the coarse and fine particulate mass concentrations. The PM_{10} mass

concentration was obtained from the sum of the coarse and fine fraction mass concentrations.

Table 1. Summary of the coarse and fine mass concentrations at Chittagong city.

Parameter	Chittagong	
	Coarse particle ($PM_{2.5-10}$)	Fine particle ($PM_{2.5}$)
Mean ($\mu g/m^3$)	24.6	45.9
Median ($\mu g/m^3$)	15.9	30.9
Standard deviation ($\mu g/m^3$)	21.9	41.2
Maximum ($\mu g/m^3$)	93.6	208
Minimum ($\mu g/m^3$)	2.07	4.18
Number of filters exposed	104	104

Table 2 shows the black carbon (BC), organic matter (OMH), and elemental concentrations and their corresponding standard deviations for both coarse and fine particulate matters. About 36% of the total coarse particulate mass is black carbon and hydrogen containing organic matter. While the black carbon and hydrogen containing organic matter accounts for more than 70% of the fine particulate mass. Thus, the fine fraction of the PM mass in Chittagong city is predominately of anthropogenic origin.

Table 2. Summary of elemental mass concentrations and BC (ng/m^3) used for RCM, PCA analysis.

Parameter	Coarse fraction		Fine fraction	
	Mean	STD	Mean	STD
OMH	5635	4695	16395	14106
BC	5644	3767	11327	4202
Na	549	636	469	560
Mg	198	163	123	283
Al	1108	1012	321	434
Si	3160	2913	861	907
S	479	217	503	678
Cl	803	648	236	355
K	574	460	1166	895
Ca	998	951	170	227
Ti	90	89	21	26
Mn	35	39	31	38
Fe	1249	1293	327	346
Cu	14	16	23	30
Zn	1240	1959	2286	3276
Br	21	15	54	56
Pb	146	205	603	810

The distribution of 24-hour average fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) mass fractions results for the Khulshi site in Chittagong is shown in Fig. 2 as monthly average box and whisker plots. The box represents 25 to 75% of the distributions of the monthly $PM_{2.5-10}$ (top) and $PM_{2.5}$ (bottom) concentrations. The horizontal bar in the box indicates the

median and (+) sign denotes the mean of the distribution for that month. The points lying outside the range defined by the whiskers (extreme events) are plotted as outlier dots.

From the box-whisker plots (Fig. 2), it is observed that in winter and early pre-monsoon, the mass concentrations remain high. With relatively low temperature in winter and pre-monsoon period, the mixing height becomes lower and the particulate matter is trapped nearer to the ground level. Moreover, because of low rainfall during this period, the particulate concentration from re-suspended particles increases.

It can be seen from plot and also from Table 1 that the standard deviations of the mass concentrations are large, since the distributions of values are positively skewed and heavily tailed. This large variability could be due to variations in emission rates and the influence of meteorology like wind speed, wind direction and mixing height.

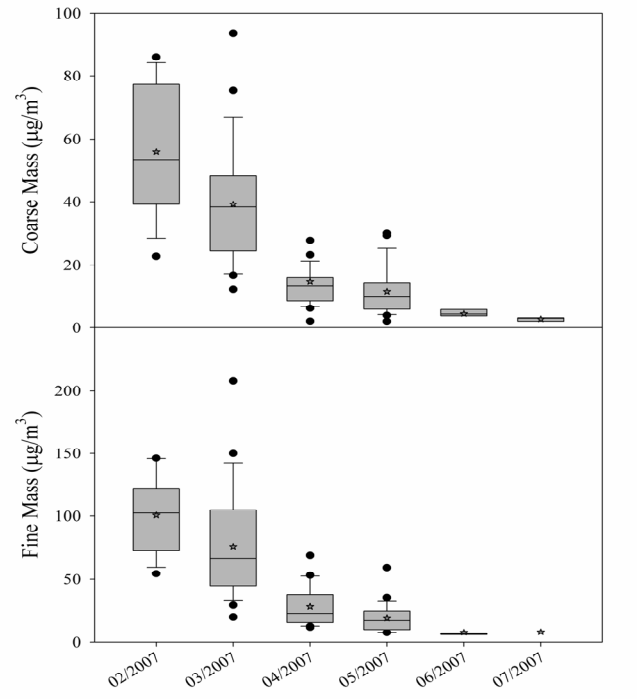

Fig. 2. Box and whisker plot for monthly mass concentration (µg/m^3) in Chittagong.

SOURCE APPORTIONMENT

The average EF values calculated for the samples collected from different sites are presented in Table 3. As a "rule of thumb", the enrichment factors greater than about 10 indicate element additions over reference levels that may be attributed to anthropogenic influences (Wiersma *et al.* 1992). Most of the elements like H, S, Cl, K, Cr, Co, Ni, Cu,

Zn, Br, Rb, Sr and Pb considered to be of anthropogenic origin have EF values more than 10. It may be concluded that these elements come from anthropogenic activities. From the correlation between elements it is possible to find out the anthropogenic sources.

Table 3. Enrichment factors of the PM samples.

Fraction	H	S	Cl	K	Ca	Cr	Co	Ni	Cu	Zn	Br	Rb	Sr	Pb
$PM_{2.5}$	323	924	448	10.6	0.84	11.4	35.1	10.7	60.2	4476	4019	56.8	15.8	7363
PM_{10}	39.3	166	491				19.2		9.7	611	527	27.3	6.5	527

Enrichment factors only identify the sources, which originate from the anthropogenic sources. From the biplots of enriched elements, it is possible to identify the sources. It was found that a good correlation exists between the elements like S vs OMH which may be the source of motor vehicle, K vs S for brick kiln, and Pb and Zn may come from galvanizing factories. The sources of Pb are batteries or paints. Tables 4 and 5 show the anthropogenic sources obtained from enrichment factor for coarse and fine fractions, respectively.

Table 4. Anthropogenic sources obtained from enrichment factor for coarse fraction.

Enriched elements		Slope	r^2	Source
OMH	S	3.62	0.322	Motor vehicle
Pb	Zn	0.699	0.184	Galvanizing factories/Zn source
Cu	OMH	0.195	0.012	CNG
Pb	Sr	0.007	0.023	Fugitive Pb
S	K	130	0.475	Brick kiln

Table 5. Anthropogenic sources obtained from enrichment factor for fine fraction.

Enriched elements		Slope	r^2	Source
OMH	S	3.18	0.427	Motor vehicle
Zn	Pb	1.38	0.1997	Galvanizing factories/Zn source
OMH	Co	0.103	0.112	CNG
Br	Pb	1.409	0.108	Fugitive Pb
OMH	K	0.0323	0.8335	Brick kiln

The variation of fine and coarse soil concentrations (Fig. 3) shows the monthly box and whisker plots for the coarse (top) and fine (bottom) soil estimates. During the winter and early pre-monsoon periods, the concentrations of soil were higher than any other periods.

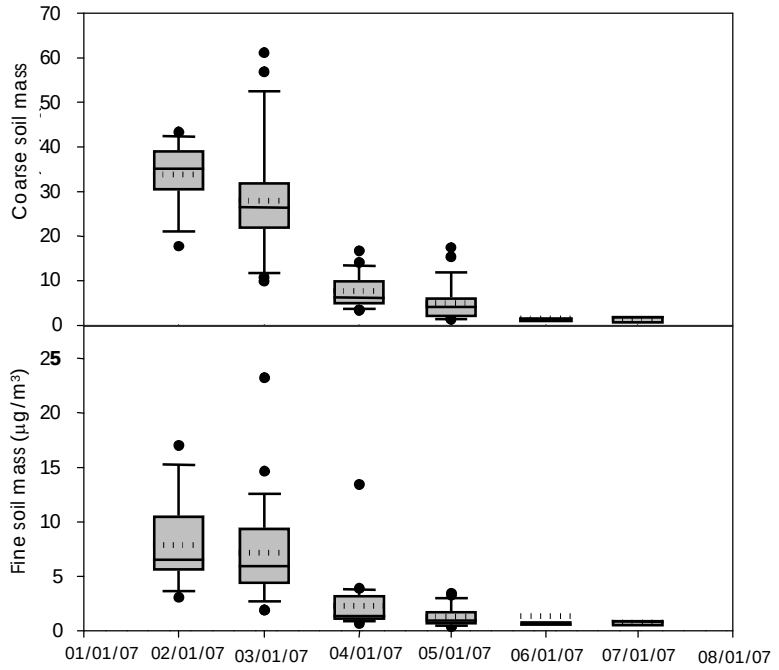

Fig. 3. Box and whisker plot for monthly soil mass concentration (µg/m^3) in Chittagong. Top: Coarse soil mass; Bottom: Fine soil mass.

The sum of the composite variables discussed in the previous section should provide a reasonable estimate of the total coarse and fine mass in comparison with the measured gravimetric mass of both coarse and fine particles on the filters. The RCM was compared with the gravimetric weight of the filters, where the least squares fit to the data gave RCM = 1.16*Weight with an R^2 = 0.88 in case of coarse mass (Figure 4, bottom) and RCM = 0.75*Weight with an R^2 = 0.82 in case of fine mass (Fig. 4, top). Table 6 gives the percentage contribution of each of RCM component in the calculated RCM values for coarse and fine particulate matter samples.

In order to produce the best possible source resolutions, it is important to have accurate and precise measurements of the particulate mass as well as determinations for as many as elements as possible to obtain better mass closure. Of the 21 elements determined in each of the 104 samples, five elements, Cr, Co, Ni, Cu, Rb and Sr, had values that were below the detection limits in over 80% of the samples. These five elements were thus eliminated from the data set for further analysis. For rest of the elements in several samples the concentration values were below detection limit. As the method cannot use missing or below detection limit data, a value is required to assign for those data values. These values were assigned a random number from a uniform random

generator to provide a number between zero and the detection limit (Hopke 1982), which gives acceptable results.

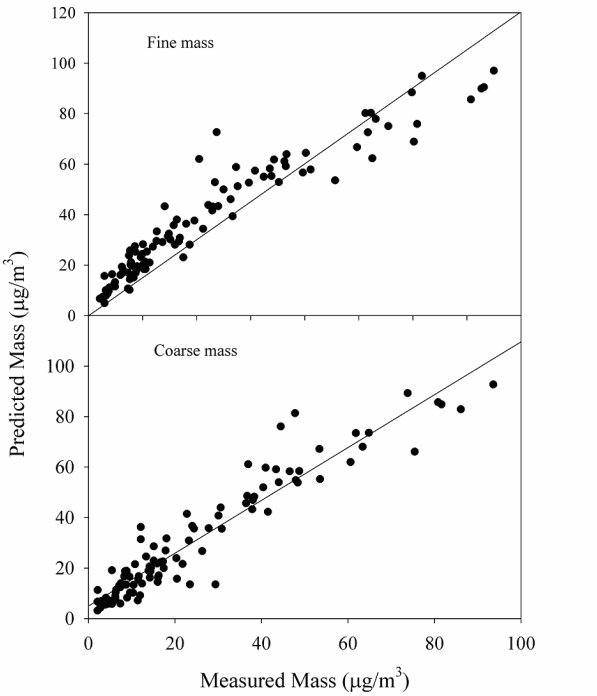

Fig. 4. The plot of RCM vs gravimetric (measured) mass. Top: Coarse mass; Bottom: Fine mass.

In order to have an unique rotation Hopke (Hopke 1982), suggested a useful empirical criterion for choosing the number of retained eigenvectors. In a number of cases of airborne particulate matter composition source identification problems, it was found that choosing the number of factors containing variance greater than one after an orthogonal rotation provided a stable solution. The total variance in each factor was calculated as the sum of the squared loading for the given factor.

Table 6. Per cent contribution of different sources of coarse and fine particulate matter from RCM calculations.

Particle size	Soil	Sulfate	Sea spray	Pb	Zn	Smoke	OMH	Black carbon
Coarse	48.6	6.34	4.46	0.45	3.98		18.1	18.1
Fine	10.2	5.37	2.70	1.53	5.92	2.47	42.4	29.3

For obtaining reliable estimates of the different sources contributing to the fine and coarse mass measured at Khulshi site, Principal Components Analysis (PCA) was used to identify major elements associated with sources. Tables 7 and 8 show the factor loadings

from the PCA analysis. Factors with two or more elements which have factor loadings above 0.3 have been highlighted. Five factors were required to explain 95% of the sample variance in case of the coarse particulate matters and six factors were required to explain 93% of the sample variance in case of the fine particulate matter. Factor loadings near 1.0 demonstrate that the element has a strong association with that individual factor.

Table 7. Principal component analysis with varimax rotation for all $PM_{2.5-10}$ data from Chittagong.

Element	Road dust	Sea salt and Zn	CNG vehicle	Motor vehicle	Fugitive Pb	Communality
OMH	0.61	0.52	0.33	0.38	0.17	0.93
BC	0.34	0.25	0.20	0.85	0.22	0.99
Na	0.25	0.91	0.09	0.10	0.19	0.94
Al	0.90	0.30	0.24	0.14	0.10	0.99
Si	0.88	0.35	0.24	0.17	0.09	0.99
S	0.87	0.08	-0.18	0.27	0.21	0.92
Cl	0.40	0.79	0.04	0.20	0.22	0.88
K	0.84	0.40	0.21	0.22	0.16	0.98
Ca	0.86	0.32	0.31	0.13	0.06	0.97
Ti	0.89	0.33	0.23	0.14	0.10	0.98
Mn	0.71	0.50	0.35	0.23	0.07	0.94
Fe	0.80	0.46	0.29	0.18	0.09	0.98
Cu	0.42	0.22	0.74	0.26	0.29	0.93
Zn	0.38	0.79	0.26	0.15	0.18	0.90
Pb	0.14	0.38	0.19	0.20	0.86	0.98
%var	74.8	9.35	5.23	3.38	2.70	95.5
Eigen	11.2	1.4	0.78	0.51	0.40	

Factor 1 in Table 7 explained 74.8% of the variance and includes high factor loadings for the elements OMH, BC, Al, Si, S, Cl, Ti, Ca, K, Mn, Fe, Cu and Zn for coarse particles and was identified as typical road dust indicators. Factor 2 explained 9.35% of the variance and was labeled sea spray/fresh sea salt which is mixed with Zn source as it include high loadings for Na, and Cl mixed with OMH, BC, Al, Si, K, Ca, Ti, Mn, Fe and Pb. Factor 3 explained 5.23% of the variance and was labeled CNG vehicle source.

Factor 4 explained 3.38% of the variance and was labeled as Motor Vehicle emissions (both diesel and gasoline engine exhaust) and shows high values of S (Begum *et al*. 2005). Factor 5 explained 2.7% of the variance and was labeled as fugitive or resuspended Pb source. Although, Pb was eliminated from the gasoline in

Bangladesh in July 1999 (Biswas *et al.* 2003), there may be substantial accumulated lead in the dust near roadways.

Table 8. Principal component analysis with varimax rotation for all $PM_{2.5}$ data from Chittagong.

Element	Road dust	Soil dust	Sea salt	Brick kiln	Motor vehicle	Fugitive Pb	Communality
OMH	0.76	0.28	0.08	0.47	–0.15	0.07	0.92
BC	0.31	0.22	0.17	0.87	0.08	0.12	0.95
Na	0.11	0.29	0.86	0.16	0.22	0.14	0.94
Al	–0.04	0.84	0.12	–0.05	0.03	0.41	0.89
Si	0.35	0.85	0.14	0.27	–0.18	–0.01	0.97
S	0.19	–0.14	0.18	0.03	0.93	–0.08	0.96
Cl	0.11	0.13	0.96	0.06	0.03	0.05	0.96
K	0.77	0.31	0.14	0.44	–0.09	0.09	0.92
Ca	0.30	0.90	0.17	0.05	–0.04	–0.07	0.94
Ti	0.25	0.89	0.12	0.24	–0.06	–0.04	0.93
Mn	0.88	0.30	0.09	0.16	0.21	0.00	0.94
Fe	0.60	0.71	0.22	0.16	–0.03	–0.06	0.95
Cu	0.91	0.14	–0.02	0.07	0.14	0.12	0.89
Zn	0.89	0.17	0.18	0.04	0.14	0.14	0.90
Pb	0.50	0.04	0.30	0.24	–0.17	0.70	0.92
%Var	52.8	14.5	11.2	6.97	4.59	3.08	93.1
Eigen	7.91	2.17	1.68	1.05	0.69	0.46	

Examining the fine particle results in Table 8, factor 1 explained 52.8% of the variance with high factor loadings for crustal elements including Si, Ti, Ca, K, Mn, Cu, Pb and Fe mixed with OMH, BC and Zn. This factor appears to be mixed with emissions from CNG vehicles as it contains high values of OMH and Cu. These elements are typical indicators of road dust. Factor 2 explained 14.5% of the variance and was labeled as soil dust that shows high values of Al, Si, K, Ca, Ti, Mn, and Fe. Factor 3 explained 11.2% of the variance and was labeled sea spray as it included high loadings for Na, and Cl. Factor 4 explained 6.97% of the variance and was labeled as brick kiln. This factor has a high value of K that most likely comes from biomass/brick kiln along with BC and OMH. Factor 5 explained 4.59% of the variance and was labeled as motor vehicle that shows high values of S (both diesel and gasoline engine exhaust). Factor 6 explained 3% of the variance and was labeled as fugitive or resuspended Pb.

CONCLUSIONS

The present study on source apportionment could resolve seven source components in fine and coarse fraction of the PM samples collected in Chittagong city. The major sources contributing to the coarse PM fraction are soil dust including resuspended soil and emissions from vehicles. Alternatively, biomass burning/emissions from brick kilns and Zn source were found to be the major contributors for fine PM. Emissions from metal work also have significant contributions to fine PM. Attention should be given to reduce contributions from those sources to improve the air quality in Chittagong city.

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