Source Apportionment of Air Particulate Matter by Chemical Mass Balance (CMB) and Comparison with Positive Matrix Factorization (PMF) Model

Bilkis A. Begum[1], Swapan K. Biswas[1], Philip K. Hopke[2*]

[1] Chemistry Division, Atomic Energy Centre, P.O. Box-164, Dhaka, Bangladesh.
[2] Center for Air Resources Engineering and Science, Clarkson University, Potsdam, NY 13699-5808, USA.

Abstract

The Chemical Mass Balance (CMB) model is one of several receptor models that have been applied to air quality management. This model quantifies contributions using chemical signatures characterizing source-types rather than contributions from individual emitters. The CMB model uses the chemical composition of ambient pollution samples to estimate the contributions of different source types to the measured pollutant concentrations. The disadvantage of the model is that it cannot separate the sources having similar chemical compositions or for those sources for which source composition profiles are unavailable. Since CMB analysis is done on a sample-by-sample basis, it is possible to estimate the daily contributions of individual sources, and can thereby provide useful information based on a limited number of samples to address air quality management issues. Samples of fine and coarse fractions of airborne particulate matter (PM) were collected using a 'Gent' stacked filter unit in two fractions of 0-2.2 μm and 2.2-10 μm sizes in a semi-residential (Atomic Energy Centre Dhaka, AECD) area from June 2001 to June 2002 of Dhaka. These samples were analyzed for elemental concentrations with PIXE. The chemical composition data set was analyzed by CMB using local source profiles obtained using a Principal Components Analysis (PCA) and regression analysis of data from this site and the source contributions are quantitatively estimated for each of the samples. The results of the CMB analysis were compared with the results obtained using positive matrix factorization (PMF) that had been done previously. It is observed that CMB provides comparable results except for limited discrepancies, especially for the $PM_{2.2}$ fractions where sources have similar elemental signatures.

Keywords: Chemical mass balance (CMB); Source; Apportionment; Positive matrix factorization (PMF); PIXE; $PM_{10-2.2}$; $PM_{2.2}$.

[*] Corresponding author. Fax : +315-268-4410

E-*mail address*: hopkepk@clarkson.edu

INTRODUCTION

Air pollution is a common concern in the large and growing metropolitan areas throughout the world, especially in South East Asian cities. There are six major pollutants termed criteria pollutants in the United States, namely, PM_{10} and $PM_{2.5}$, NO_x, SO_2, O_3 and CO (Akhter *et al.,* 2004) that have known severe health implications (Dockery *et al.,* 1994, Pope, 2000, Schwartz, 2001). Particulate matter (PM) is recognized as the most important air pollutant in the Dhaka, the capital city of Bangladesh (Begum *et al*., 2006a). Dhaka (latitude 23.77N, longitude 90.38E) is congested with a large number of motor vehicles, including both public and private transportation. The vehicles are believed to constitute the dominant source of air pollution in Dhaka (Begum *et al.,* 2004). Moreover, construction of roads and buildings is taking place continuously throughout the city. Many small-scale factories are also located in and around the city. Brickfields that use coal and wood to fire the bricks, have grown up around Dhaka city because of the increased demand of construction materials and these brickfields are also major contributors to the air pollution in winter since they operate only winter when the meteorological conditions are sufficiently dry (Salam *et al.,* 2003).

The identification of various sources of airborne particulate matter (APM) and assessment of their impact on the aerosol composition of an air shed are major goals of contemporary atmospheric research. The national air quality standards for the APM in many countries around the world have also created the need to identify the particle sources so that effective control strategies can be designed and implemented. The initial efforts at the identification of particle sources focused on dispersion models of point sources, and in most cases, this approach resulted in substantial reduction in APM levels. Thus, additional methods were required to identify and quantitatively apportion the particle mass to sources with better resolution. These methods are called receptor models. Measured properties of collected ambient samples are used in these models to infer the contributions of the sources to the pollutant concentrations. This approach requires that the samples be collected at the location of interest, the receptor site and that the samples so collected be analyzed for the properties that are characteristic of the pollutant sources. Therefore, measurements of the elemental composition of atmospheric aerosol particles are needed for compliance monitoring, studies of environmental deposition, and source attribution by receptor modeling techniques (Gordon, 1988).

Receptor models have been developed to identify and apportion the contributions of various sources to the airborne particulate matter concentrations. Factor analysis (FA) techniques are multivariate data analysis methods that are used in environmental studies to estimate the number and compositions of the sources as well as their contributions to the samples taken at the receptors. Principal Components Analysis (PCA) is one of the common forms of FA (Jollife, 1986). This method extracts the principal components, explaining the majority of variance of the

data matrix that are then qualitatively interpreted as possible sources (Wolff *et al.,* 1985). However, this method suffers from several drawbacks (Paatero *et al.,* 1993). A new approach to FA called Positive Matrix Factorization (PMF) was developed (Paatero, 1997; 1999). This model uses a weighted least-square approach and imposes non-negativity constraints for fitting the FA model. Fairly large data sets with reasonable uncertainties are needed both in PCA and PMF models in order to produce good results. However, obtaining such data sets is often difficult and expensive. Moreover, it may be required to differentiate source contributions in different seasons or during specific limited duration pollution episodes. In such cases, large data set may not be available.

The Chemical Mass Balance (CMB) model has the capability of analyzing individual samples if local source profiles or fingerprints are known (Abu *et al.,* 2002). In order to obtain a source profile of a given particle source, two things must be determined: first, which chemical species (elements, compounds, or ions) characterize that source profile and second, the relative proportions of each of those elements that provide the signature for the source profile at a sampling site. These fingerprints or profiles can then be used in the CMB model (Watson *et al.,* 1990) together with the sample's species concentrations and their associated analytical errors. In an earlier study, it was found that the source profiles obtained from PCA were comparable with PMF (Begum *et al.,* 2006b), so the source profiles obtained from the PMF analysis could be used as input to the CMB model. Since the CMB analysis could be applied even on a single sample, it is possible to estimate the daily contribution from the individual source if the source profiles are properly estimated. It is also applicable when only a small data set is available (Chowdhury *et al.,* 2003). Profiles obtained from a large set of data from one location could be applied to a small data set from another site if it is known that the same or similar sources will affect both sites. Particularly if these profiles can be derived easily from readily available data sets (Begum *et al*., 2006a).

Particulate matter (PM) is found to be the major pollutant of concern in Dhaka and from the time series pattern, it is observed that PM concentration depends on the prevailing meteorological conditions (Begum *et al.,* 2006a). Thus, the contributions of the sources should be estimated for the different seasons and these contributions could be useful for supporting specific policy interventions to reduce emissions.

The main objective of this study is to analyze elemental and BC concentration data of PM samples collected at a semi-residential site at Dhaka using the CMB model to apportion the source contributions. The local source profiles used in this analysis were obtained from a previous data analysis study (Begum *et al.,* 2006b) and the data set used for the analysis is from July 2002 to June 2003 of the same site. The results of the CMB analyses have been compared with those obtained from the PMF analysis performed in an earlier study (Begum *et al.,* 2004) to ascertain if such an approach of using derived profiles can be applied to a data set to obtain good

mass apportionments. As a result of this apportionment, the seasonal patterns of various pollutant sources are also discussed.

MATERIAL AND METHODS

Sampling

Sampling was done using a "Gent" type stacked filter sampler (Hopke *et al.,* 1997) capable of collecting particulate matter in the $PM_{10-2.2}$ (coarse) and $PM_{2.2}$ (fine) size fraction samples collected between June 2001 and June 2002. The samples were collected on an 8 μm pore filter (coarse fraction) and on a 0.4 μm pore filter (fine fraction). The sampler was placed on the flat roof of the building in the AECD area. The roof height is 5 m and the intake nozzle of the sampler was located 1.8 m above the roof. The intake was about 80 m away from the roadside. The sampler was placed so that the airflow was unobstructed. The effective sampling time was varied between 6-20 hours distributed uniformly over 24 hours in a day to avoid filter clogging and so that the flow rate remained within the prescribed limits of the sampler. This flow control ensures proper size fractionation and collection efficiency.

PM mass and BC determination

The masses of the coarse and fine fraction samples were determined by weighing the filters before and after the exposure. A Po-210 (alpha emitter) electrostatic charge eliminator (STATICMASTER) was used to eliminate the static charge accumulated on the filters before each weighing.

The concentration of black carbon (BC) in the fine fraction of the samples is determined by reflectance measurement using an EEL-type Smoke Stain Reflectometer. Secondary standards of known black carbon concentrations are used to calibrate the Reflectometer (Biswas *et al.,* 2003).

Multielement analysis

Multielement analyses of the samples collected during above cited time intervals were made using Proton Induced X-ray Emission (PIXE) at the Institute of Geological and Nuclear Sciences (IGNS), New Zealand. 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 Micrometer thin target standards in the similar experimental conditions. Data on the concentrations of eighteen elements, black carbon and mass were available for further analysis.

Meteorological conditions

In Bangladesh, the climate is characterized by high temperatures, high humidity most of the year, and distinctly marked seasonal variations in precipitation. Based on the meteorological conditions, the year can be divided into four seasons, pre-monsoon (March-May), monsoon (June-September), post-monsoon (October-November) and winter (December-February) (Salam *et al.,* 2003). The meteorological data used in this study was obtained from a local meteorological station, located about 5 kilometers north of the semi-residential site.

Data analysis by Chemical Mass Balance

The CMB model uses the chemical composition of airborne particulate matter samples to estimate the contributions of different source types to the measured concentrations at the receptor site using previously estimated source fingerprints. The chemical composition of each source-type's emission (commonly known as source profile or source fingerprints) may be estimated from PCA or sometimes a reasonable estimate may be obtained from the United States Environmental Protection Agency (USEPA) library (SPECIATE). In the present study, the source profiles have been estimated from the results obtained from prior analysis (Begum *et al.,* 2006b). This model quantifies contributions from chemically distinct source-types rather than contributions from individual pollution emitters. This model cannot separate the sources that have very similar chemical compositions. It does estimate the source contributions for each individual sample. The particle composition may differ from one sample to the next owing to differences in emission rates, wind directions, wind speed, and changes in emission compositions. However, the variations in the source compositions are not taken into account in these analyses.

The model consists of the following equations

$$C_i = F_{i1}S_1 + F_{i2}S_2 + F_{i3}S_3 + ... + F_{ij}S_j + .. + F_{iJ}S_J \qquad \text{i = 1…I, j = 1…J} \qquad (1)$$

where C_i = Concentration of species I measured at a receptor site

F_{ij} = Fraction of species i emissions from source j

S_j = Estimate of the contribution of source j

I = Number of chemical species

J = Number of source types

These equations have a unique solution only when the number of species is equal to or greater than the number of sources. It has been generally found that the greater number of species, the more precise the apportionment. The method also requires precision estimates for the C_i and F_{ij}

values as model input. These precision estimates result in realistic uncertainties associated with the source contribution estimate, S_j that are calculated by the model.

Conditional Probability Function (CPF)

To analyze point source impacts from various wind directions, the conditional probability function (CPF) (Kim *et al.,* 2003a; 2003b) as calculated using source contribution estimates coupled with wind direction values measured on site. To minimize the effect of atmospheric dilution, daily fractional mass contribution from each source relative to the total of all sources was used rather than using the absolute source contributions. The same daily fractional contribution was assigned to each 3 hours period of a given day to match to the 3 hours average wind direction. Specifically, the CPF is defined as

$$CPF = \frac{m_{\Delta\theta}}{n_{\Delta\theta}} \tag{2}$$

where $m_{\Delta\theta}$ is the number of occurrence from wind sector $\Delta\theta$ that exceeded the threshold criterion, and $n_{\Delta\theta}$ is the total number of data from the same wind sector. In this study, $\Delta\theta$ was set to be 45 degrees. The threshold was set at the upper 50 percentile of the fractional contribution from each source. The sources are likely to be located to the directions that have high conditional probability values.

RESULTS AND DISCUSSION

Table 1 shows the average mass black carbon and elemental concentrations along with median and standard deviations of fine and coarse particulate samples collected at this semi-residential area (SR) site in Dhaka, Bangladesh. Here all of the samples are collected on weekdays. The yearly average values for both PM_{10} and $PM_{2.2}$ masses (Begum *et al.,* 2006a) are much higher than the 1997 USEPA standards as well as the Bangladesh national air quality standard for PM_{10} and $PM_{2.5}$ that were set at 50 μg/m^3 and 15 μg/m^3, respectively. The PM_{10} mass concentrations are also higher than the WHO guideline range (60-90 μg/m^3).

CMB analysis was performed using the CMB7 software obtained from the USEPA website (Watson *et al.,* 1990). Local source profiles obtained from PCA (Begum *et al.,* 2006b) method, except road dust, metal smelter and biomass burning source, are provided as the input of the model and the source contributions are quantitatively estimated for each of the samples. The profiles are different for coarse and fine particulate matter. PCA and PMF method can produce comparable source profiles, The PMF source profiles have been used for these sources. The source profiles for coarse and fine fractions are presented in Figs. 1 and 2. The CMB model

diagnostics for PM are in the acceptable limits as prescribed in the CMB user guidelines (Watson *et al.,* 1990). For each of the PM samples, the R^2 was between 0.8 and 0.9 and the chi-square varied from 0.89-4. Eight sources could be identified for both coarse and fine fraction of the PM samples collected from semi-residential area of Dhaka. Figs. 3 and 4 present the measured mass concentrations of the individual samples compared with the predicted mass concentrations by the CMB model. The R^2 values are 0.81 and 0.72 for coarse and fine particulate matter samples, respectively. In the case of the fine particle mass (Fig. 4), the CMB could account for about 86% of the measured mass concentrations. However, rather poorer reproduction of the mass concentrations was obtained for the coarse particle samples (64%). It is entirely clear why so much less mass was apportioned for the coarse particle samples as compared to the fine particle samples. There are no measurements of the carbonaceous constituents of the coarse PM that would include primary biological materials such as plant parts and broken pollen grains. The small amount of potassium they contain would be taken up by the biomass burning factor. Thus, some contribution from primary biological components would appear to be part of the low mass apportionment.

Table 1. Mean, median and standard deviations of PM mass, BC and elemental concentration at a semiresidential area (concentrations in ng/m^3).

Parameter	Fine particulate matter			Coarse particulate matter		
	204 samples			204 samples		
	Mean	Median	S.D.	Mean	Median	S.D.
Mass	22250	16800	15411	42876	32460	37094
BC	7887	7067	3952			
Na	592	552	285	477	355	523
Mg	433	403	228	919	866	412
Al	502	413	331	2148	1869	1228
Si	792	599	648	4459	3840	2732
P	277	220	204	641	516	413
S	1292	1107	776	1223	874	936
Cl	140	111	106	827	481	933
K	390	323	218	747	574	524
Ca	163	116	166	1385	1190	818
Ti	16.9	12.4	17.9	150	116	105
Cr	7.99	6.85	7.76	25.2	11.7	127
Mn	8.87	8.16	5.57	30.7	27.6	18.8
Fe	207	186	132	1306	1185	766
Ni	3.21	2.81	2.34	4.58	3.37	3.51
Cu	5.05	4.70	4.07	13.2	7.64	43.4
Zn	272	189	247	375	281	367
Br	10.6	5.45	7.87	17.4	12.4	28.5
Pb	164	58.1	557	124	37.0	350

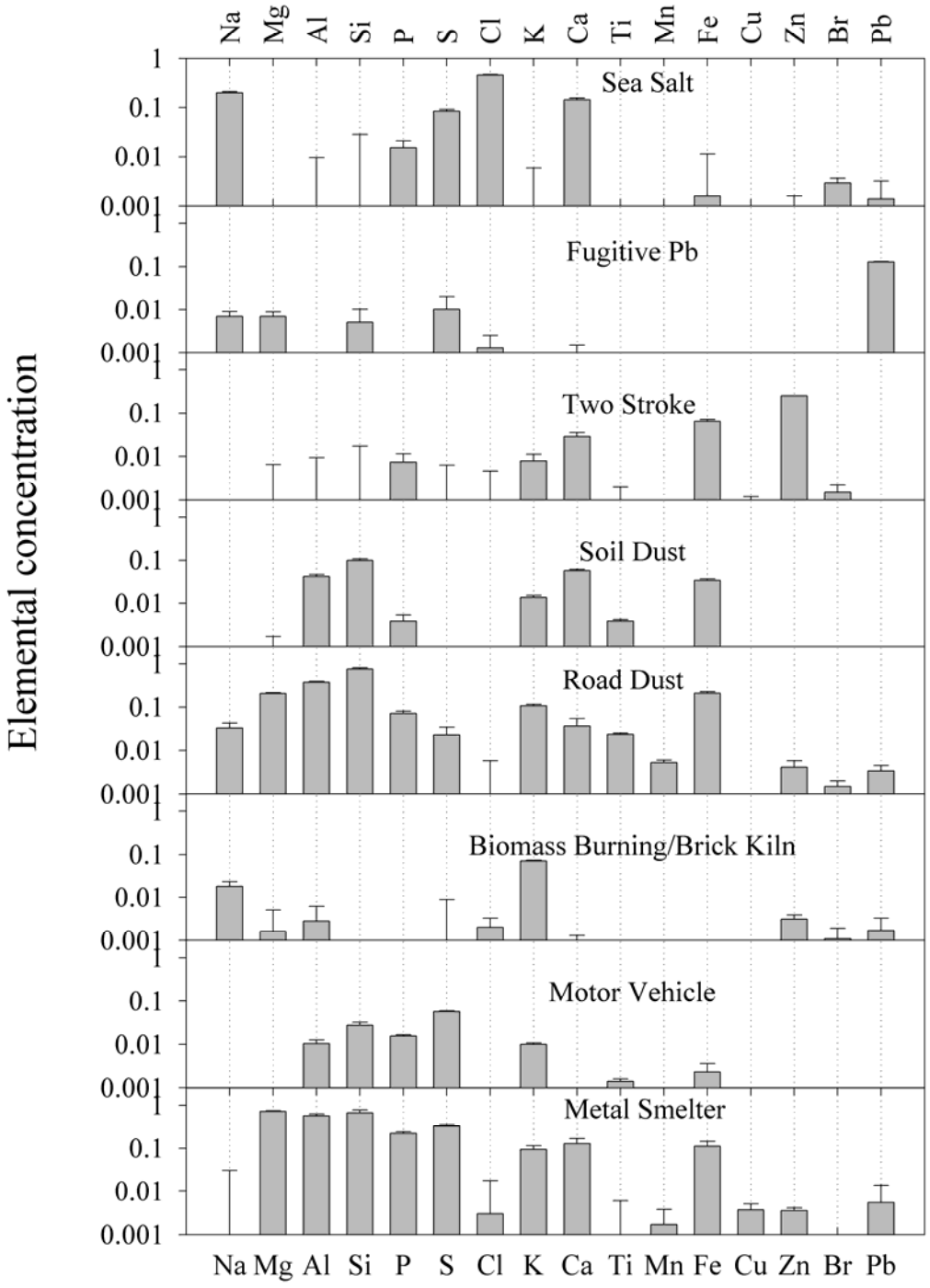

Fig. 1. Source fingerprint for coarse particulate matter at semi-residential area in Dhaka

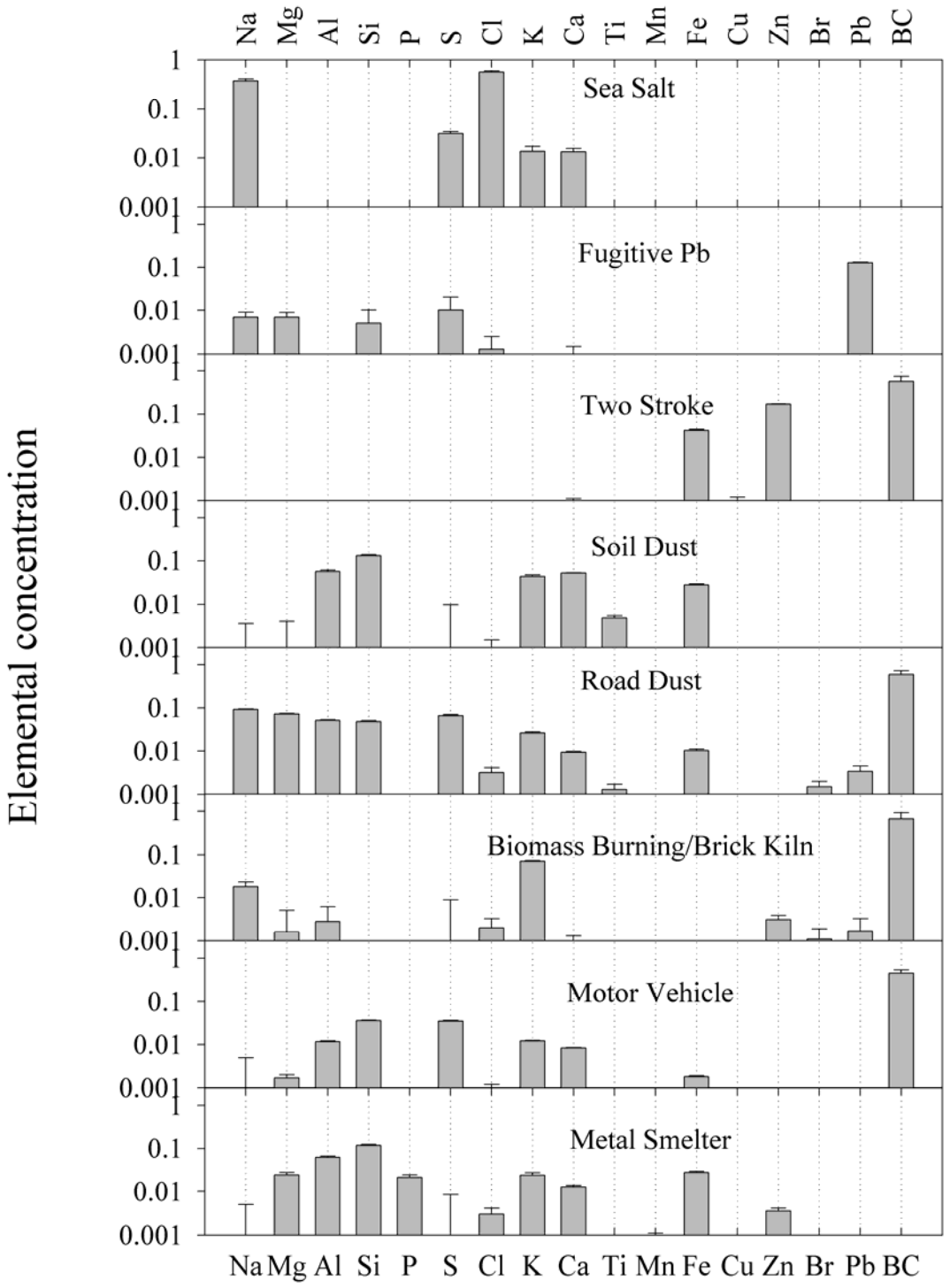

Fig. 2. Source fingerprint for fine particulate matter at semi-residential area in Dhaka

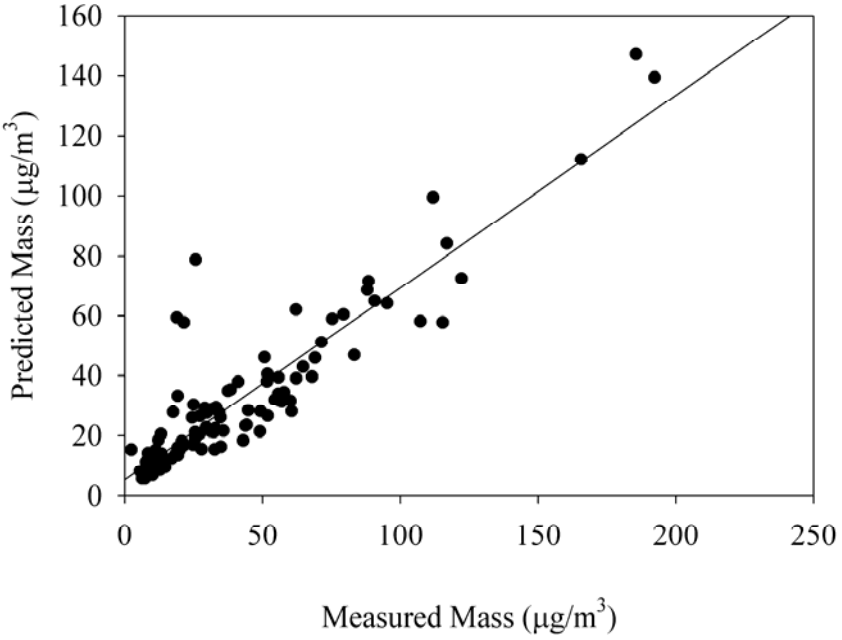

Fig. 3. Regression between measured and predicted mass concentrations for coarse particulate matter.

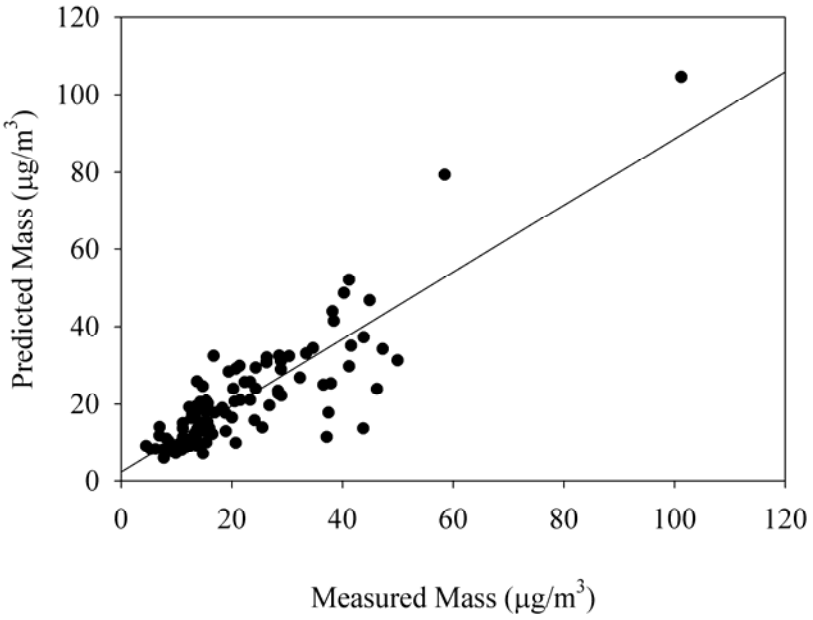

Fig. 4. Regression between measured and predicted mass concentrations for fine particulate matter.

The eight sources resolved by CMB modeling contributing to the both the fine and coarse fractions of the PM samples were attributed to sea salt, two-stroke engine, soil dust, road dust, motor vehicle, metal smelter, fugitive/resuspended Pb and brick kiln. The resolved time series for these contributions are presented in Figs. 5 and 6. Figs. 7 and 8 shows polar plots of the

conditional probability function for each of the resolved sources. It can be observed from the CPF plots that the sources have same wind directional patterns as obtained by PMF modeling (Begum *et al.,* 2004). The CMB modeling could resolve two additional sources, fugitive Pb and a brick kiln source, that the previous PMF modeling was unable to derive.

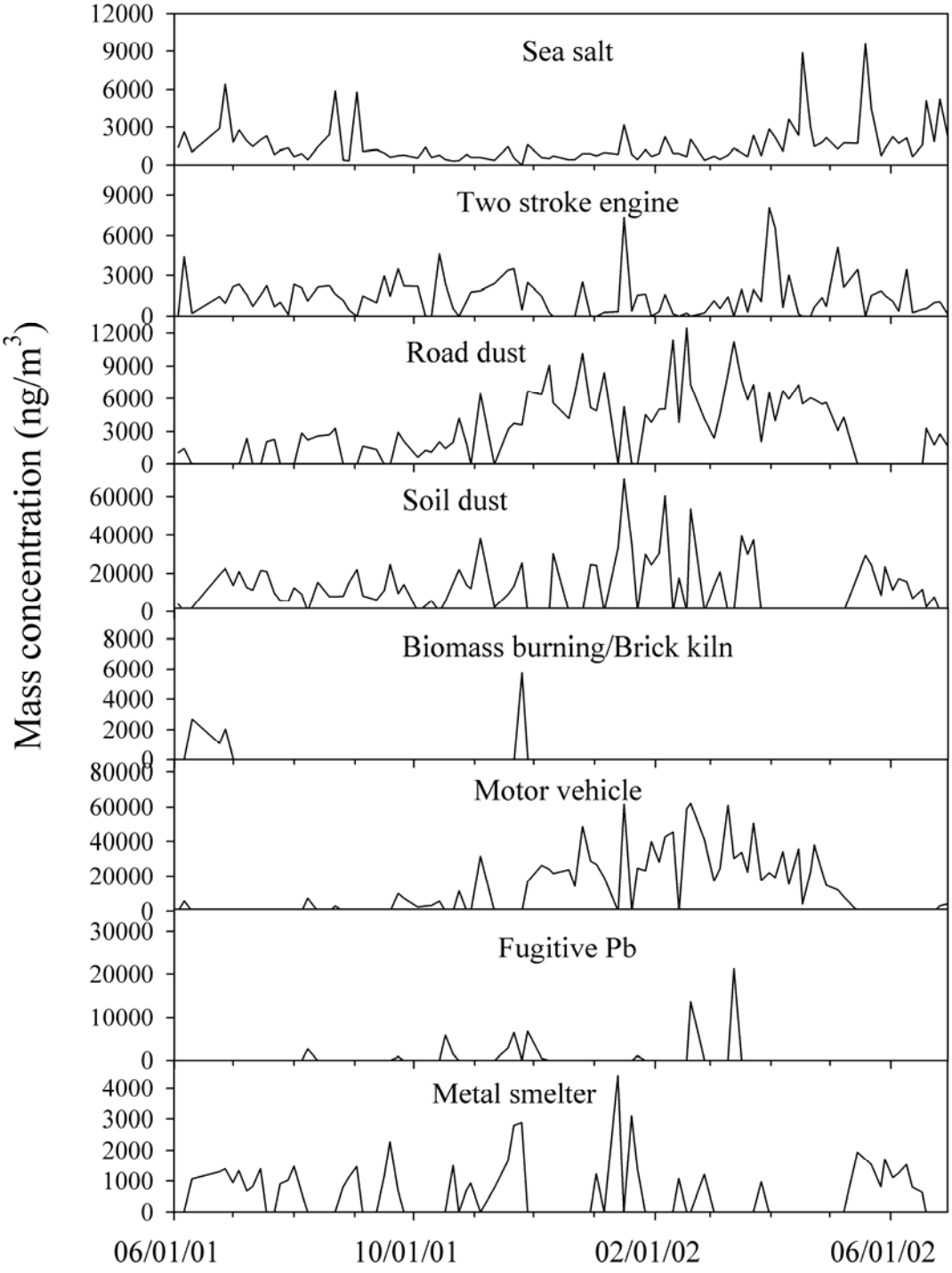

Fig. 5. Time variations for coarse particulate matter at the semi-residential site in Dhaka.

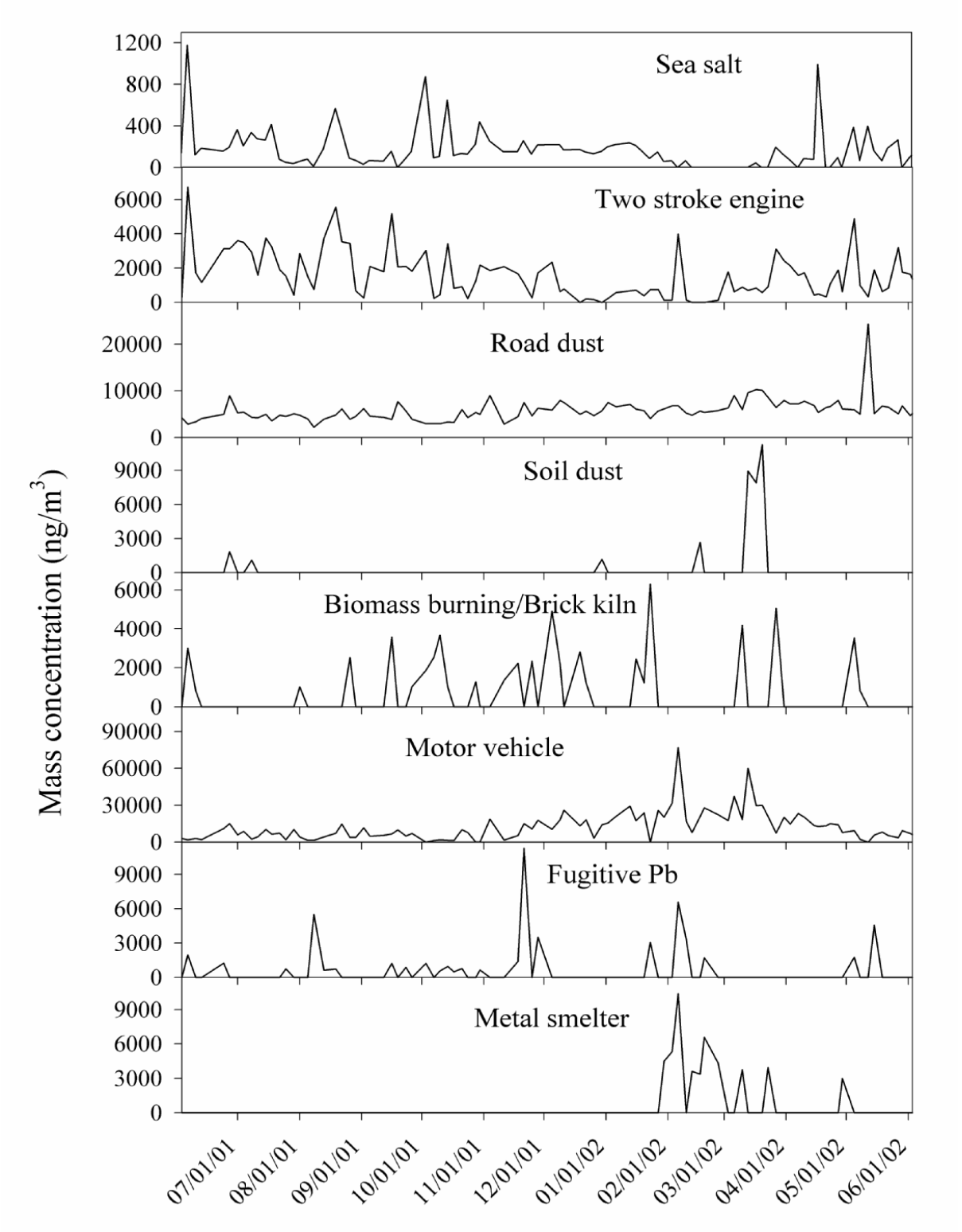

Fig. 6. Time variations for fine particulate matter at the semi-residential site in Dhaka.

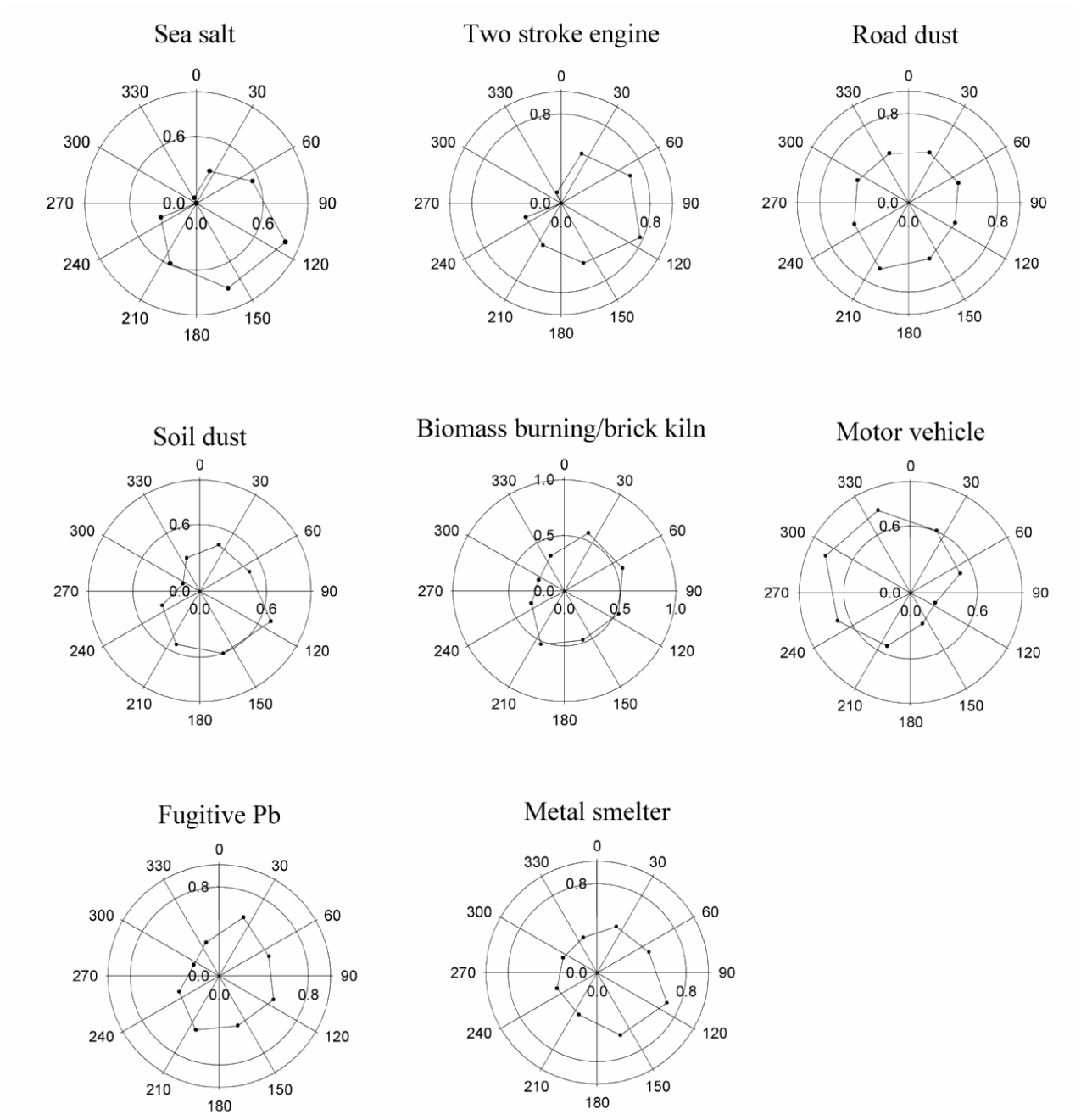

Fig. 7. The relationship of source contributions with wind direction for coarse particulate matter at the semi-residential site.

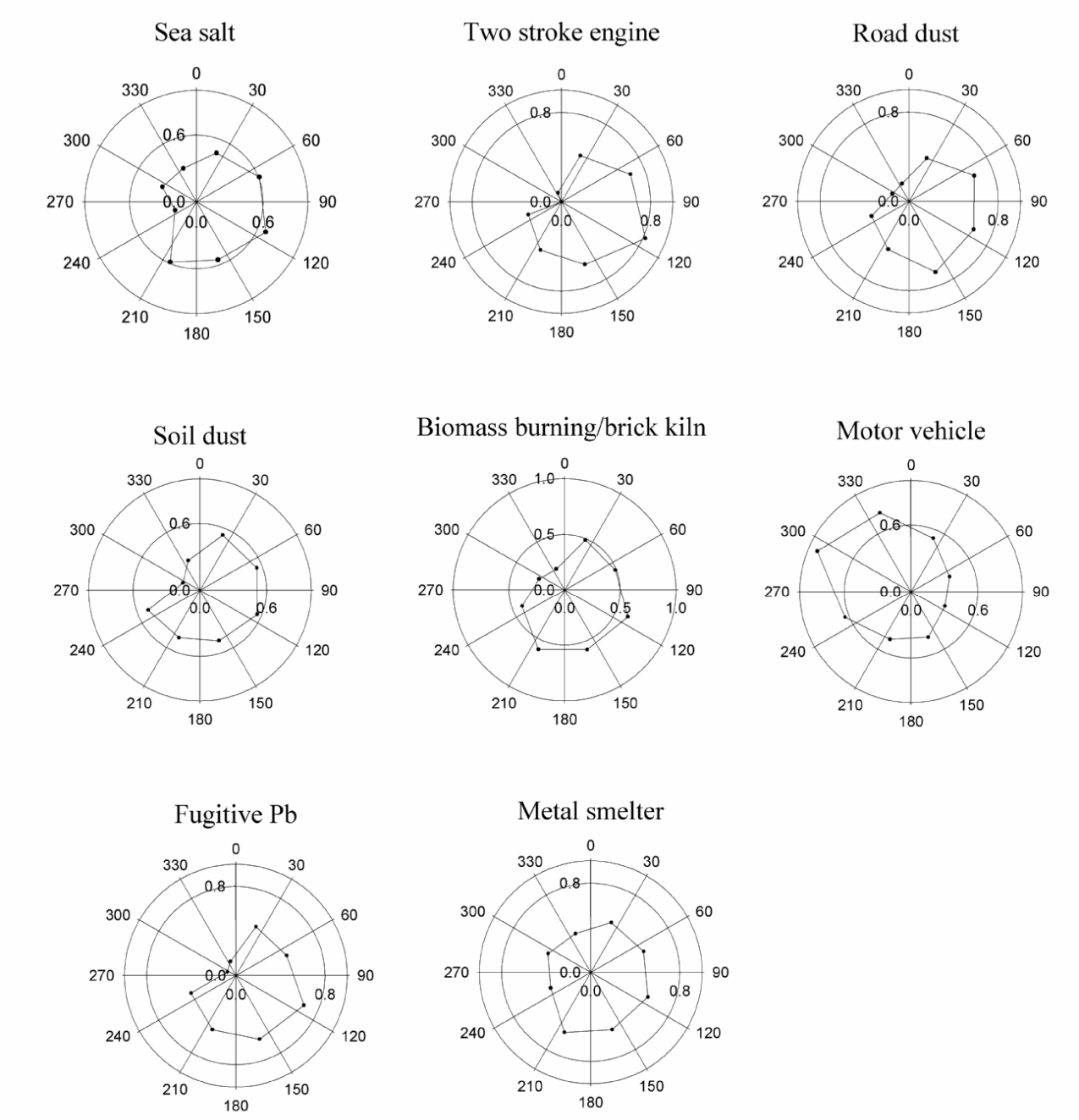

Fig. 8. The relationship of source contributions with wind direction for fine particulate matter at the semi-residential site.

SOURCE RELATED CLASSIFICATION OF THE ELEMENTS

Soil dust and road dust

The soil contains the characteristic elements Al, Si, K, Ca, Ti and Fe (Watson *et al.,* 2001a; 2001b). It has a few high peaks during winter season when rainfall is at a minimum. The CPF plots show the likely direction of the source. Wind blown dust could produce these particles.

The road dust is characterized by the Al, Si, Ca, K, Fe, Na, Mg, P, S, and BC and this source did not varied much during the study period. This profile contains S and BC as well as other soil components (Bhave *et al.,* 2001, Ho *et al.,* 2003). These species are attributed primarily to diesel vehicle emissions. The source is influenced by southerly winds. There is a busy road about a kilometer away on the eastern side of the sampling site with heavy traffic. This road probably explains the dependence of the source on wind direction.

Motor vehicles

Historically, lead has been used as the most reliable tracer of traffic. However, the gradual shift from leaded to unleaded petrol as fuel for automobiles has reduced traffic emissions of this element (Biswas *et al.,* 2003). The motor vehicle (mixed diesel and gasoline engine exhaust) source profile is characterized by the high BC and S (Maricq *et al.,* 2002; Kim *et al.,* 2003b; Begum *et al.,* 2004). This source is also mixed with the crustal elements, Mg, Al, Si, and Fe, suggesting the vehicular exhaust is mixed with resuspended road dust. This factor has a higher contribution during the dry season and is influenced by northwesterly wind. In Dhaka, two main traffic corridors extend in this direction. The sampling site lies between two large bus stations; one is on the northern side and the other station is to the west of the site.

Two-stroke engine emissions

The factor has the characteristic of having high Zn, BC and Fe (Chueinta *et al.,* 2000). These elements are characteristic of lubricating oil used in two-stroke engine where engine oil is mixed with fuel. Zinc compounds are extensively employed as lubricants, antioxidants and as detergent/dispersant improvers for lubricating oils. Oxidation of lubricating oils upon exposure to air at high temperature results in the formation of metal oxide particles. The oil may also contain metal wear particles consisting of Zn, Cu, and Cd-bearing alloys (Drew, 1975). Zn could also be contributed from galvanized materials, tire wear and the use of zinc compounds in rubber production (Fergussion *et al.,* 1991). This factor profile includes Na, S, and Mn suggesting that part of this factor may be attributed to refuse burning normally used in the slums for cooking purposes. The two-stroke engines emit large quantities of organic carbon (Snow and Crews, 2004). However, organic carbon was not measured in this study. These engines also emit black smoke and BC was part of the profiles. The time series plot shows relatively higher contributions

in the wet season (Morales *et al.,* 1996) and the CPF plot shows the influence of easterly and southeasterly winds (Begum *et al.,* 2004). There are some galvanizing industries situated in the south and southeastern parts of Dhaka. Moreover, a relatively large, lower income population (slum dweller) lives in this area.

Sea salt

High Na and Cl characterize the sea salt component in the fine PM. The time series plot shows random peaks mostly during monsoon period between June and October and dominated by easterly winds. The sea salt component was expected to be seen during the monsoon season, when the wind is normally from the southeast.

Biomass burning/brick kiln

The third source may represent biomass burning/brick kiln characterized by K (Azad *et al.,* 1998; Watson *et al.,* 2001a; Begum *et al.,* 2004). The source has seasonal variation as indicated in time series plot with high contributions in the dry season. The CPF plot shows a stronger influence of both southwesterly and northeasterly wind (Begum *et al.,* 2004). There are many brick kilns around Dhaka and a large amount of wood and coal are burnt in those kilns. The kilns normally operate only between October and April and thus, explain the higher source contribution during the winter. In addition, slums in and around the city use biomass as a cooking fuel.

Metal smelters

The source characterized by high Al, Mg, Si, Ca and mixed with K, Fe, Mn, Zn, etc. could represent metal smelting sources (Harris *et al.,* 2005). The time series plot shows random peaks mostly between November and February. This factor was influenced mostly by easterly and southeasterly wind. There are a significant number of small industrial facilities involved in metal working including foundries distributed around Dhaka.

Fugitive Pb

The fourth source shows a high value of Pb together with crustal elements (Na, Mg, Si, P, S and Cl) indicating fugitive/resuspended Pb (Harris *et al.,* 2005). Pb has generally shown a tendency to be associated more with the fine fraction than the coarse fraction (Young *et al.,* 2002). This tendency is because the Pb particles were formed at high temperature as part of the vehicle exhaust. However, handling of lead batteries in preparation for the reclamation of the lead produces coarse particle lead (Glover *et al.,* 1991). The time series plot shows that this source contains several peaks all over the sampling period in case of fine particulate matter. The CPF plot shows the dependence on southeasterly winds and thus, battery recycling could again be a

significant contributor to this factor. Although, Pb was eliminated from the gasoline in Bangladesh beginning in July 1999, there may be substantial accumulated lead in the dust near roadways because lead has a very long residence time in surface soil (Young *et al.,* 2002). There are also Pb-battery recycling plants in old town of Dhaka in this direction. In addition, the use of lead weights to balance tires leads to their loss from the wheels into the roadways where they can be broken down over time to produce coarse lead particles.

Comparison of CMB and PMF results

Table 2 presents the comparison of estimated average source contribution obtained from CMB model calculations and those obtained from PMF modeling for the coarse and fine particulate matter using the same chemical composition data sets of all the samples. Both the modeling technique could produce comparable results, especially for coarse PM. In case of the CMB analysis, soil dust together with road dust accounted for about 47.6% of the total coarse PM whereas from PMF analysis sum of these factors were just over 50%. The second largest contributor was motor vehicle emissions including two-stroke engine exhaust that was about 43.3% and PMF also resolved that source as a 44% contribution to the mass. As CMB analysis is done on a sample-by-sample basis, it could be found that there are only few days where the contribution of fugitive Pb at the receptor site was high. Pb may come from the paints and the batteries industries. CMB resolved about 5% sea salt from the coarse fraction while PMF produced a somewhat smaller value.

Table 2. The source contribution obtained from CMB.

Source	From CMB	From PMF	From CMB	From PMF
	Coarse particle		Fine particle	
Sea salt	5.04	4.45	0.80	1.00
Two stroke	4.35	3.78	7.36	9.36
Soil dust	38.0	43.0	1.61	10.2
Road dust	9.61	7.30	27.0	19.4
Motor vehicle	38.9	40.2	55.3	38.2
Metal smelter	1.76	1.21	2.25	9.96
Fugitive Pb	1.93		2.67	
Brick kiln	0.35		2.97	11.9

In case of fine PM, several differences could be noted. Although CMB and PMF calculation produced different source contribution for soil and road dust, the sum of the contributions of these sources remained the same in both sets of results. Similarly, the CMB analysis accounted for higher motor vehicle emissions and lower metal smelter, fugitive Pb and brick kiln contributions than the PMF model calculations.

In the CMB modeling, if sources have similar chemical signatures, it may not be able to differentiate between these sources. The collinearity between source profiles may account for the differences in calculated source contributions in the two modeling techniques. For example, sea salt and two-stroke sources have unique source fingerprints and thus, both models found similar results. However, in the case of the dust sources, more ambiguity in the results were obtained although the total dust contributions were quite similar.

CONCLUSIONS

From CMB calculations, eight sources have been identified from both coarse and fine particulate matter and this modeling technique produces comparable source contribution. Since the CMB analysis is done on a sample-by-sample basis, it is possible to determine the daily contribution from the individual source if the source profiles are properly estimated. This feature could result in the separation of more sources in the CMB results than in the PMF modeling. Although Pb was eliminated from the gasoline in Bangladesh in July 1999, source apportionment using CMB model calculation shows that about 3% of the contribution to FPM is from fugitive component containing Pb that is comparable with the PMF result of hot spot site, Dhaka (Begum *et al.,* 2005). There may be substantial accumulated Pb in the dust near roadways. There could also be fugitive Pb emission from battery and other industries.

The sample-by-sample approach in CMB also can provide useful information based on limited number of samples collected at a receptor site to address air quality management issues where PMF cannot be used. As particulate matter (PM) is major pollutant of concern in Dhaka and from time series pattern, it is observed that PM concentration depends on prevailing meteorological conditions, the contributions of sources on monthly average could also be estimated (Figs. 9 and 10) and in turn could be useful for undertaking policy interventions. The method can also be helpful in identifying pollution episodes which occurs for a short period.

ACKNOWLEDGEMENTS

The work is financially supported partly by the RCA/IAEA through the research project RAS/7/013 and the Ministry of Science and Information & Communication Technology (MOSICT), Government of the People's Republic of Bangladesh under the special grant. The authors thankfully acknowledge those assistances. The authors would like to acknowledge Dr. Andreas Markwitz and Dr. W.J. Bill Trompetter of Institute of Geological and nuclear sciences for providing the accelerator facilities for PIXE analysis and useful discussion during analysis.

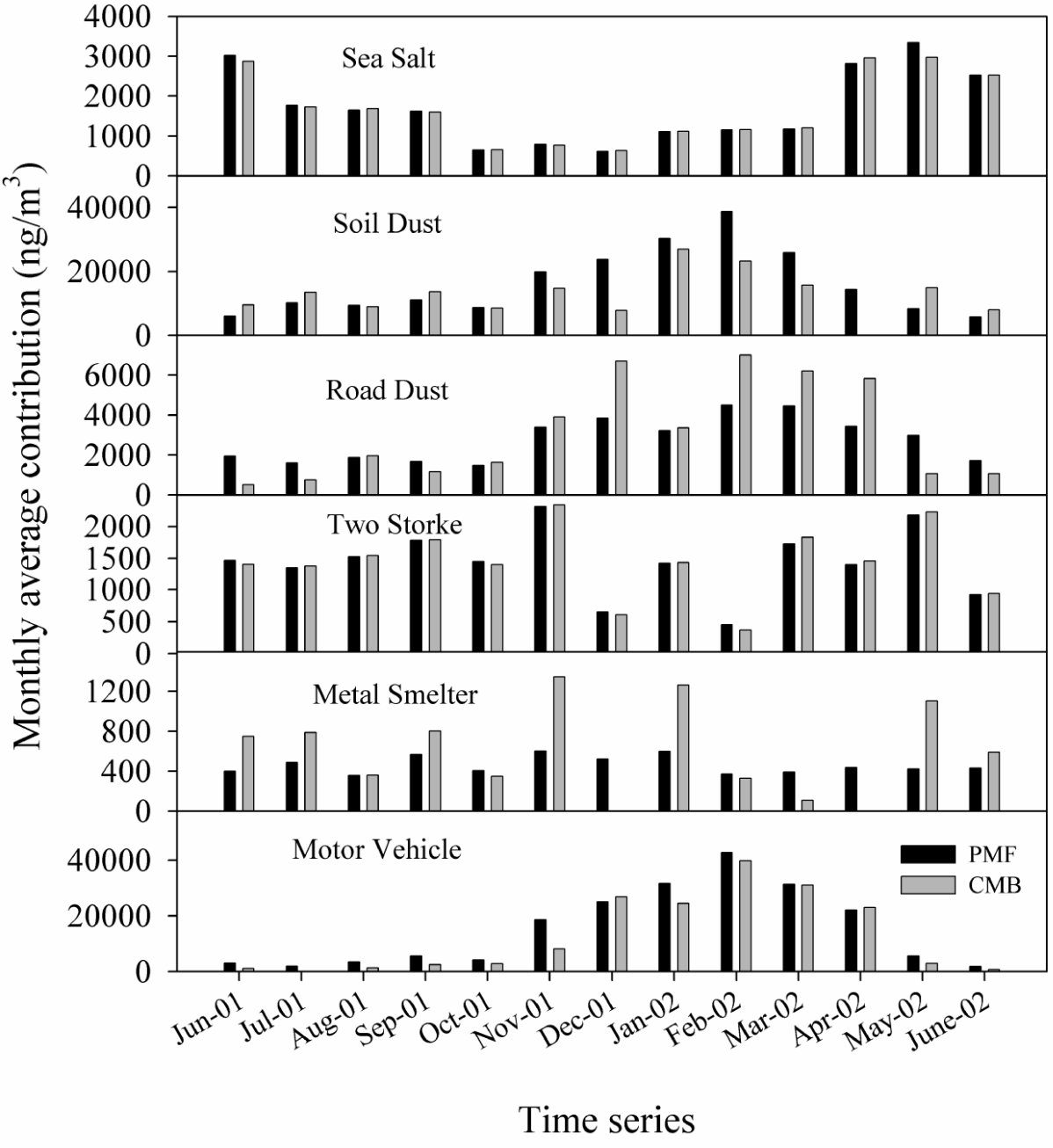

Fig. 9. Monthly average contribution of sources obtained from PMF and CMB methods during the study period for coarse particulate matter at semi-residential site in Dhaka.

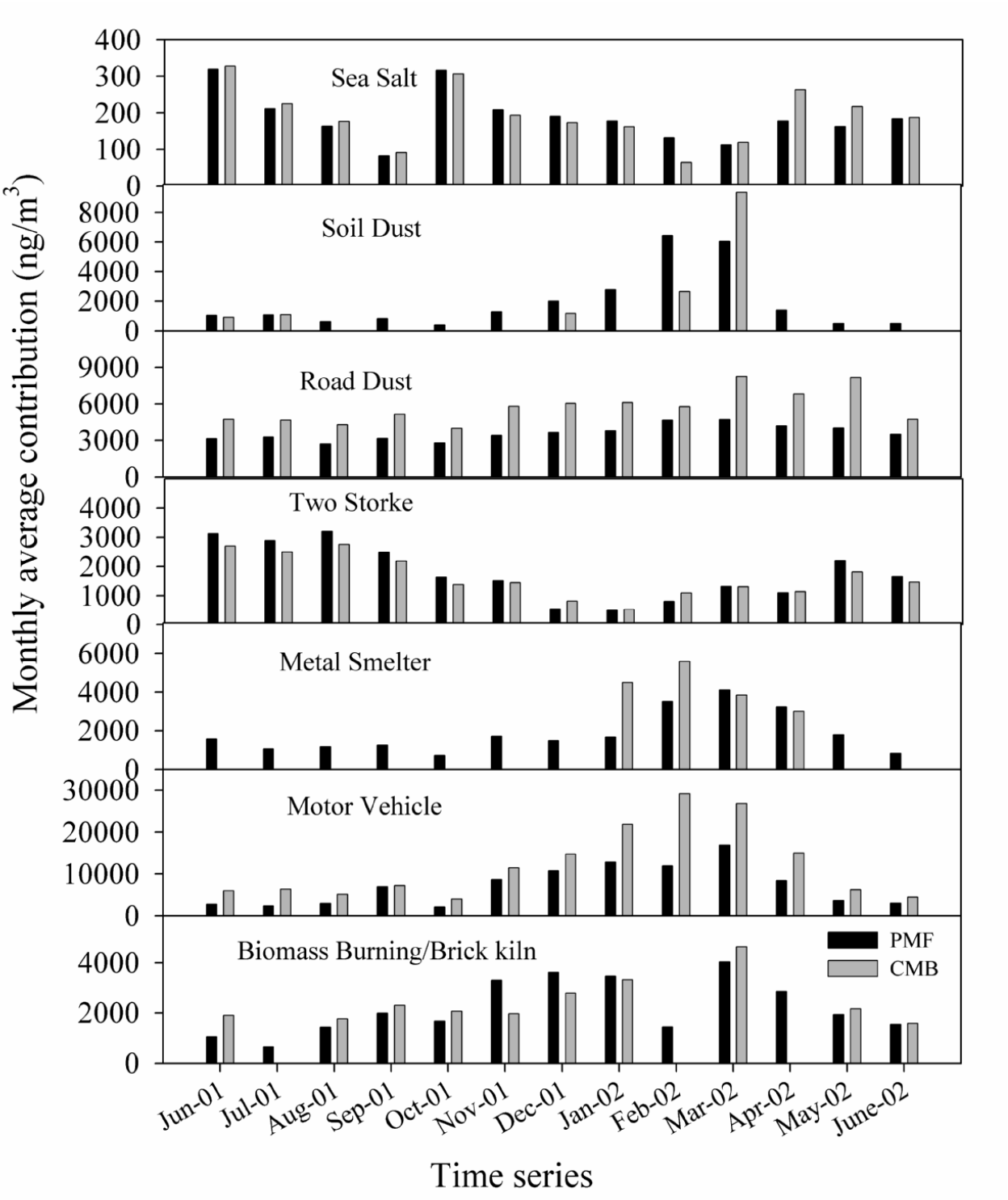

Fig. 10. Monthly average contribution of sources obtained from PMF and CMB methods during the study period for fine particulate matter at semi-residential site in Dhaka.

REFERENCES

Abu-Allaban, M., Gertler, A.W. and Lowenthal, D.H. (2002). A Preliminary Apportionment of the Sources of Ambient PM_{10}, $PM_{2.5}$ and VOCs in Cairo. *Atmos. Environ.* 36: 5549-5557.

Akhter, S., Islam, M.A., Hossain, A.M.S., Quadir, S.M.A., Khan, A.H., Begum, B.A., Khaliquzzaman, M. and Swapan, K.B. (2004). Air Quality Monitoring Program in Bangladesh: Trends analysis of Criteria Pollutants and Source Apportionment of Particulate Matter in Dhaka, Bangladesh. *Clean Air Initiative (Asia),* www.baq2004.org.

Azad, A.K. and Kitada, T. (1998). Characteristics of the Air Pollution in the City of Dhaka, Bangladesh in Winter. *Atmos. Environ.* 32: 1991-2005.

Begum, B.A., Biswas, S.K. and Hopke, P.K. (2006a). Temporal Variations and Spatial Distribution of Ambient $PM_{2.2}$ and PM_{10} Concentrations in Dhaka, Bangladesh. *Sci. Total Environ*. 358: 36-45.

Begum, B.A., Biswas, S.K., Hopke, P.K. and Cohen, D.D. (2006b). Multi-Element Analysis and Characterization of Atmospheric Particulate Pollution in Dhaka. *Aerosol Air Qual. Res*. 6: 334-359.

Begum, B.A., Biswas, S.K., Kim, E., Hopke, P.K. and Khaliquzzaman, M. (2005). Investigation of Sources of Atmospheric Aerosol at a Hot Spot Area in Dhaka, Bangladesh. *J. Air Waste Manage. Assoc*. 55: 227-240.

Begum, B.A., Kim, E., Biswas, S.K. and Hopke, P.K. (2004). Investigation of Sources of Atmospheric Aerosol at Urban and Semi-Urban Areas in Bangladesh. *Atmos. Environ.* 38: 3025-3038.

Bhave, P.V., Fergenson, D.P., Prather, K.A. and Cass, G.R. (2001). Source Apportionment of Fine Particle Matter by Clustering Single-Particle Data: Tests of Receptor Model Accuracy. *Environ. Sci. Technol*. 35: 2060-2072.

Biswas, S.K., Tarafdar, S.A., Islam, A., Khaliquzzaman, M., Tervahattu, H. and Kupiainen, K. (2003). Impact of Unleaded Gasoline Introduction on the Concentration of Lead in the Air of Dhaka, Bangladesh. *J. Air Waste Manage. Assoc*. 53: 1355-1362.

Chowdhury, Z., Zheng, M. and Russell, A. (2003). Source Approtionment of the Ambient Fine Particles in Mumbai, Delki and Kolkata, *Georgia Institute of Technology*, World Bank Report (ESMAP, 281/04).

Chueinta, W., Hopke, P.K. and Paatero, P. (2000). Investigation of Source of Atmospheric Aerosol Oa Urban and Suburban Residensial Areas in Thailand by Positive Matrix Factorization. *Atmos. Environ.* 34: 3319-3329.

Dockery, D.J. and Pope, C.A. (1994). Acute Respiratory Effects of Particulate Air Pollution. *Ann Rev Public Health*. 15: 107-132.

Drew, H.M. (1975). Metal-Based Lubricant Compositions. *Noyes Data Corporation, NJ*.

Fergussion, J.E. and Kim, N.D. (1991). Trace Elements in Street and House Dust: Source and Speciation. *Sci. Total Environ*. 100: 125-150.

Glover, G.E., Hopke, P.K., Vermette, S. J., Landsberger, S. and D'Auben, D. R. (1991). Source Apportionment with Site Specific Source Profiles. *J. Air Waste Manage. Assoc*. 41: 294-305.

Gordon, G.E. (1988) Receptor Models. *Environ. Sci. Technol*. 22; 1132-1142.

Harris, A.R. and Davidson, C.I. (2005). The Role of Resuspended Soil in Lead Flows in the California South Coast Air Basin. *Environ. Sci. Technol*. 39: 7410-7415.

Ho, K.F., Lee, S.C., Chow, J.C. and Watson, J.G. (2003). Characterization of PM_{10} and $PM_{2.5}$ Source Profiles for Fugitive Dust in Hong Kong. *Atmos. Environ.* 37: 1023-1032.

Hopke, P.K., Xie, Y., Raunemaa, T., Biegalski, S., Landsberger, S., Maenhaut, W., Artaxo, P. and Cohen, D. (1997). Characterization of the Gent Stacked Filter Unit PM_{10} Sampler. *Aerosol Sci. Technol*. 27: 726-735.

Jollife, I.T., *Principal Component Analy* (Springer-Verlag, New York, 1986).

Kim, E., Hopke, P.K. and Edgerton, E.S. (2003a). Source Identification of Atlanta Aerosol by Positive Matrix Factorization. *J. Air Waste Manage. Assoc*. 53: 731-739.

Kim, E., T.V. Larson, P.K. Hopke, C. Slaughter, L.E. Sheppardand Claiborne, C. (2003b). Source Identification of $PM_{2.5}$ in an Arid Northwest U.S. City by Positive Matrix Factorization. *Atmos Research*. 66: 291-305.

Maricq, M.M., Chase, R.E., Xu, N. and Laing, P.M. (2002). The Effects of the Catalytic Converter and Fuel Sulfur Level on Motor Vehicle Particulate Matter Emissions: Light Duty Diesel Vehicles. *Environ. Sci. Technol*. 36: 283-289.

Morales, J.A., Pirela, D. and Duran, J. (1996). Determination of the Levels of Na, K, Ca, Mg, Fe, Zn and Cu in Aerosols of the Western Venezuelan Savannah Region. *Sci. Total Environ*. 180: 155-164.

Paatero, P. (1999). A Table-driven Least Squares Program for Solving Multilinear Problems, Including th N-Way Parallel Factor Analysis Model. *J. Comput. Graph. Stat.* 8: 854-888.

Paatero, P. (1997). Least Squares Formulation of Robust Non-Negative Factor Analysis. *Atmos. Environ.* 37: 23-35.

Paatero, P. and Tapper, U. (1993). Analysis of Different Methods of Factor Analysis as Least square Fit Problem. *Atmos. Environ.* 18: 183-194.

Pope, C.A. (2000) Epidemiology of Fine Particulate Air Pollution and Human Health: Biologic Mechanisms and Who's at Risk. *Environ. Health Perspect*. 108: 713-723.

Salam, A., Bauer, H., Kassin, K., Ullah, S.M. and Puxbaum, H. (2003). Aerosol Chemical Characteristics of a Mega-City in Southeast Asia (Dhaka, Bangladesh). *Atmos. Environ.* 37: 2517-2528.

Schwartz, J. (2001). Is There Harvesting in the Association of Airborne Particles with Daily Deaths and Hospital Admissions *Epidemiology*. 12: 55-61.

Watson, J.G. and Chow, J.C. (2001a). $PM_{2.5}$ Chemical Source Profiles for Vehicular Exhaust, Vegetation Burning, Geological Materials and Coal Burning in Northwestern Colorado during 1995. *Chemosphere*. 43: 1141-1151.

Watson, J.G. and Chow, J.C. (2001b). Source Characterization of Major Emission Source in the Imperial and Maxicali Valleys along the US/Mexico Boarder. *Sci. Total Environ*. 276: 33-47.

Watson, J.G., Robinson, N.F., Chow, J.C., Henry, R.C., Kim, B., Nguyen, Q.T., Meyer, E.L. and Pace, T.G. (1990). Receptor Model Technical Series, Vol. III, CMB7 User's Manual, http://www.epa.gov/ttn/chief/software.htmlUS Environmental Protection Agency, Report EPA-450/4-90-004, January.

Wolff, G.T., Korsog, P.E., Kelly, N.A. and Fermam, M.A. (1985). Relationships between Fine Particulte Species, Gaseous Pollutants and Meteorological Parameters in Detroit19. *Atmos. Environ.* 19: 1341-1349.

Young, T.M., Heeraman, D.A., Sirin, G. and Ashbaugh, L.L. (2002). Resuspension of Soil as a Source of Airborne Lead near Industrial Facilities and Highways. *Environ. Sci. Technol.* 36: 2484-2490.

Received for review, May 1, 2007
Accepted, July 6, 2007