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Identification and Apportionment of Sources from Air Particulate Matter at Urban Environments in Bangladesh

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Executive Summary

Particulate air pollution is the major concern in four major cities, Rajshahi, Dhaka, Khulna and Chittagong, in Bangladesh and thus it is necessary to understand the characteristics of the pollutant as well as sources for further improvement of the air quality. In this view, particulate matter (PM) sampling was done between September 2010 to July 2012 from four Continuous Air Monitoring Stations (CAMS) located at Farmgate in Dhaka, Sapura in Rajshahi, Baira in Khulna and at Khulshi in Chittagong. PM sampling was performed using dichotomous samplers, which collect samples in two size fractions: $PM_{2.5}$ and $PM_{2.5-10}$. All the samples were analyzed for mass, black carbon (BC), delta-C and elemental compositions. The data sets for each site were analyzed for sources with receptor modelling (PMF2). The identified sources include brick kilns, soil dust, road dust, motor vehicle, metal smelter, fugitive Pb, Zn source and sea salt sources in case of coarse particles $PM_{2.5-10}$ depending on site. Among them, more than 62% of the $PM_{2.5-10}$ was soil and road dust in Rajshahi, Chittagong and Khulna sites but in Dhaka, the dust contribution was about 38%. For fine particles $PM_{2.5}$, the identified sources are similar to those for the coarse particle samples, but their contributions are different. It was found that more than 63% of the fine particle mass comes from anthropogenic sources such as brick kilns, wood burning, biomass burning, and motor vehicles. The contribution of mass as well as black carbon and delta-C from the motor vehicles is much less than from brick kilns or biomass burning sources. The Government of Bangladesh is trying to reduce the emission from brick kilns by adopting green technologies for brick production. There is also long-range transport of fine particles during winter time. The impacts of local sources also increase due to poorer dispersion conditions in the winter, and the absence of precipitation. Hence, in order to reduce the local particulate pollution, it will be necessary to take policy actions locally, as well as regionally.

Identification and Apportionment of Sources from Air Particulate Matter at Urban Environments in Bangladesh

1 Introduction

This study is the first preliminary analysis of PM and BC data under Task 3 of the Bangladesh Air Pollution Studies (BAPS) project. The basis of Task 3 work is outlined in the report Task 3 (Source Apportionment) Inception Report prepared by NILU June 2012 [31].

Currently, urban air pollution and its effects are an issue of great concern for developing countries. To address the air pollution issues, it is important to know the possible sources and their strengths so actions can be taken that can effectively and efficiently improve air quality. Local sources can be controlled by local initiatives, but regional as well as trans-boundary issues would require intergovernmental interventions.

Several studies have shown that $PM_{2.5}$ has significant negative impact on human health [1-3]. Previously, source apportionment studies were performed for the same site in Dhaka city using data from June 2001 to June 2002 [4-6]. These source apportionment studies found that vehicles emissions were normally responsible for about 50% of fine particles ($PM_{2.5}$ particles) in Dhaka. Coarse particles ($PM_{2.5-10}$ particles) mainly originate from mechanical processes [7]. During that period, gasoline and diesel were mainly used as fuel to run motorized vehicles. The Bangladesh government has enacted a number of policies to reduce the concentration of ambient particulate matter during 2001 to 2003. Source apportionment based on the PM data can be used to examine the effect on these policy implementations. Several studies [6-8] have shown that there is also trans boundary contribution during the wintertime when wind blows from north and northwest directions [9].

The Department of Environment (DoE) conducted a 23-month air quality monitoring program at Continuous Air Monitoring stations (CAMS) in four major cities, Rajshahi, Dhaka, Khulna and Chittagong beginning in September 2010. In this study, samples were collected at these stations in these four different cities using dichotomous samplers. These samples were analyzed for their compositions. The resulting data were analyzed using receptor modelling (Positive Matrix Factorisation; PMF2) for source apportionment in order to examine if there are significant changes in the source characteristics raising from the policy interventions.

2 Materials and Methods

2.1 Sampling

Samples were collected on 37 mm diameter Teflon filters using Thermo Andersen dichotomous samplers, which were programmed to sample at 16.7 lpm for proper size fractionation. The samplers at each station were positioned with the intake upward and located in an unobstructed area at least 30 cm from any obstacle to air flow. The sampler inlets were placed at a height of 10 m above ground level. Appropriate QA/QC protocol was followed by DoE technicians during sampling and mass measurements. Quality assurance of the sampling was ensured by using appropriate laboratory and field blanks. The sampling protocol was every third day starting from September 2010 and continuing to July 28, 2012 at essentially all sites. After sampling, the filters were brought to the conditioned weighing room of DoE directly from the sampling site for equilibration and PM mass measurement. Care was taken in transporting the exposed filters, so that there should be no PM loss.

2.2 Site description and measurement period

The capital city Dhaka is congested with a large number of motor vehicles, including both public and private vehicles. Many small factories are also located in and around the city. The CAMS-2 site is at Farm Gate in Dhaka (latitude: 23.76°N; longitude: 90.39°E). Farmgate is characterized as a hot spot site due to the proximity of several major roadways, intersections and large volumes of vehicles the area [5]. The site is surrounded by commercial and semi industrial areas. It was found from the previous source apportionment study that the main pollutant source categories are road dust, soil dust, sea salt, Zn source, motor vehicle and brick kiln in this site [10].

Chittagong (latitude 22.22°N, longitude 91.47°E) has the largest port in Bangladesh and has heavy motor vehicular traffic, especially the central city area covering about 10 km^2. The main road network in the city runs from the port area northward towards the industrial areas. These roads are also heavily trafficked with persistent traffic jams most of the day. Trucks transporting goods between the port and the industrial areas constitute a significant part of the traffic, and the combination of the hilly nature of the area, the stop and go mode of the congested traffic and the age and heavy loading of most of the trucks causes large emissions of black diesel smoke. A Continuous Air Monitoring Station (CAMS) is operated in Chittagong to measure criteria pollutants. The location of the CAMS-3 is at the Chittagong Television Station Campus at Khulshi, which is on a hilltop about 2.5 km northwest of the Chittagong downtown area and about 100 meters above the surrounding area. The location is not strongly affected by nearby air pollution sources, and it is considered representative of the urban background air pollutant concentrations of the city [11]. The major source categories were biomass burning/brick kilns, soil dust, road dust, Zn source (including two-stroke motorcycles), motor vehicles, CNG vehicle, and sea salt in the Chittagong aerosol [11].

Rajshahi, a metropolitan city, is situated in the northern region of Bangladesh (latitude 24.37°N, longitude 88.70°E) and near the border with India. The location of the CAMS-4 is in Sapura at the Divisional Forest Office. There are a few small industries surrounding the sampling site. The climatic conditions are very similar to Dhaka. Since there are limited industries apart from brick kilns in Rajshahi city, it has been found that the contribution of biomass burning at this site is highest [4]. This biomass burning contribution may originate from the brick industry, domestic burning/residential combustion (cooking with low grade fuels), or from transboundary transport.

Khulna, the third largest city of the country, is situated in the southern region of Bangladesh (latitude 22.48°N, longitude 89.53°E) and near the Bay of Bengal. Being located in a large river delta, it is the second port area of Bangladesh. The CAMS station, CAMS-5, is located at Samagic Bonayan Nursery and Training Center in Baira, which is about 3 km north of Khulna main town. There are many small factories near the sampling site (both west and south sides), which are producing touchwood, a special type of fuel, which is made by rice husk and used as fuel for cooking. The PM samples were collected between September 2010 and July 2012 depending on the cities (Table 1).

Table 1: Summary of PM, BC and Delta-C concentrations (µg/m3) included in the modeling.

	Rajshahi (24.38ᴼN, 88.61ᴼE)				**Dhaka (23.76ᴼN, 90.39ᴼE)**				**Khulna (22.48ᴼN, 88.61ᴼE)**				**Chittagong (22.36ᴼN, 91.80ᴼE)**			
	PM 2.5-10	**PM 2.5**	**BC**	**Delta-C**	**PM 2.5-10**	**PM2.5**	**BC**	**Delta-C**	**PM 2.5-10**	**PM2.5**	**BC**	**Delta-C**	**PM 2.5-10**	**PM2.5**	**BC**	**Delta-C**
Min	2.36	14.4	2.06	0.41	3.88	15.5	1.05	0.05	3.36	6.20	1.44	0.01	2.15	9.34	0.57	0.09
Max	283	471	46.1	26.1	207	171	17.2	8.96	208	179	14.6	4.74	135	211	13.0	7.33
Mean	88.2	149	13.0	5.69	63.9	63.5	7.11	2.96	45.7	59.4	5.60	1.57	40.7	73.3	7.59	3.41
STD	58.1	96.8	7.12	3.92	40.4	37.6	3.24	1.74	33.5	43.9	2.93	1.07	31.7	50.7	3.45	2.05
Median	74.7	117	10.8	4.90	55.1	56.0	7.33	2.93	41.9	50.2	5.26	1.46	33.9	74.2	8.47	3.59
Sample size	213	206			187	175			146	136			122	114		
Sampling period	01/09/2010 to 31/07/2012				23/08/2010 to 01/07/2012				16/09/2010 to 23/02/2012				03/12/2010 to 29/02/2012			

2.3 PM mass and BC analysis

PM mass was measured in the laboratory of the DoE. The $PM_{2.5}$ masses were determined by weighing the filters before and after exposure using a microbalance [12]. The filters were equilibrated for 24 h at a constant humidity of 45% and a constant temperature (22degC) in the balance room before every weighing. A Po-210 (alpha emitter) electrostatic charge eliminator was used to eliminate the static charge accumulated on the filters before each weighing. The difference in weights for each filter was calculated and the mass concentrations for each $PM_{2.5}$ and $PM_{2.5\text{-}10}$ samples were determined.

Black carbon (BC) measurements were conducted with a two-wavelength transmissometer (model OT-21, Magee Scientific, Berkeley, CA). The two-wavelength transmissometer measures the optical absorption of the ambient PM sample at 880 nm (BC) and 370 nm (UVBC) [13]. Organic components of wood combustion particles have enhanced optical absorption at 370 nm relative to 880 nm. A calculated variable, Delta-C signal (UVBC(370nm) – BC(880nm)), has been suggested as an indicator of wood combustion particles, but is not a direct quantitative measurement of their mass concentrations [14-15].

2.4 Multi-elemental Analysis

Multi-elemental analyses of the collected samples were made using X-Ray Fluorescence (XRF) using a Spectro X-LAB2000 spectrometer at Clarkson University, USA. Twenty six species determined in all including black carbon (BC) and delta-carbon (Delta-C) for each size fraction of the 342 samples. Eight elements (P, Sc, V, Ni, Ga, Ge, Rb, and Sr) had missing or below detection limit values for more than 80% of the cases and were eliminated from the data analyses. Concentration data for twenty chemical species (Na, Mg, Al, Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, As, Se, Br, and Pb, BC, Delta-C) and mass concentration were available. Organic carbon was not measured.
The data quality of the available elemental concentration together with mass, BC and delta-C were tested by a reconstructed mass (RCM) analysis comparing the computed RCM values with the gravimetric weight of the filters [7]. The least squares fit to the data were compared with the measured mass in order to check the data quality (Table 2). It was found that due to the missing of organic carbon, the least square fitting in case of the fine particle samples was not as good as for the coarse particle samples.

Table 2: Summary of the least squares fit from RCM calculation to the measured mass in each site

Site	Coarse		Fine	
	Slope	***Coefficient***	***Slope***	***Coefficient***
Rajshahi	0.90	0.80	0.71	0.39
Dhaka	0.82	0.77	0.59	0.32
Khulna	0.66	0.62	0.56	0.52
Chittagong	0.91	0.88	0.81	0.49

2.5 Meteorological Conditions

In Bangladesh, the climate is characterized by high temperatures and high humidity for most of the year, with distinctly marked seasonal variations in precipitation. According to 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) [16]. The winter season is characterized by dry soil conditions, low relative humidity, scanty rainfall, and low northwesterly prevailing winds. The rainfall and wind speeds become moderately strong and relative humidity increases in the pre-monsoon season when the prevailing wind direction changes to southwesterly (marine). During the monsoon season, wind speeds increase further, and the air mass becomes purely marine in nature. In the post- monsoon season, the rainfall and relative humidity decrease, as does the wind speed. The wind direction starts shifting back to northeasterly [17]. The meteorological data used in this study were obtained from a local meteorological station, located about 2 kilometers north of the CAMS in Dhaka.

2.6 Back Trajectory Calculation

Using models of atmospheric transport, a trajectory model calculates the position of the air being sampled backward in time from the receptor site for those days when the concentration was high. The trajectories are presented as a sequence of latitude and longitude values for the endpoints of each segment representing each specific time interval being modeled. The vertical motion of air parcels is considered during this model. The NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT-4) [18] model was used to calculate the air mass backward trajectories. Archived REANALYSIS meteorological data were used as input. The trajectories were computed backward in time up to 120 hours (5 days). Tick marks on the trajectory plots indicate 6-hour movement locations.

2.7 Positive Matrix Factorization Modeling

PMF is a source-receptor model that solves the equation:

$$x_{ij} = \sum_{i=1}^{p} g_{ik} f_{kj} + e_{ij} \qquad (1)$$

where x is the matrix of ambient data collected at the receptor site, consisting of the species starting from Na, Mg, Al, Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Pb, BC and Delta-C in columns and dates in rows, g is the matrix of source contributions, where each source k contributes to each sample *i*, and *f* is the mass of each element *j* in each source *k* [19-21]. The best solutions were found to be seven and six factors in Rajshahi, seven factors in case of Dhaka, six factors in case of Khulna and seven factors for elemental compositions of the coarse and fine particulate matter fraction respectively. Details of this model are described elsewhere [4]. PMF2 has the ability to handle the incomplete data such as missing data, below detection limit data and negative values after blank correction by giving low weights to such data points. In this work, any missing data were replaced by the geometric mean of corresponding elements. Half of the detection limit was used for any value below detection limit and its uncertainty was set to 5/6 of detection limit value [22].

The other important feature for this analysis was using FPEAK to control rotations in PMF2. By setting positive value of FPEAK, the routine is forced to subtract the F factors from each other yielding more physically realistic solutions [23]. An additional approach, called "G space plotting" for PMF modeling [24] was utilized to explore the rotational ambiguity. This idea derives from the concepts of edges representing correlation in the results. The G space plotting helps to identify the edges that show the factors that are "independent" in the factor analysis. The rotation can then be controlled by FPEAK until an appropriate distribution of the edges is achieved. The summaries of regression slopes and coefficients from PMF modelling are presented in Table 3. The detailed description of factor profile is described in Result and discussion (Section 3). The PMF solution was evaluated by comparing the predicted mass of both coarse and fine fractions (sum of the contributions from resolved sources) with measured mass concentrations.

2.8 Conditional Probability Function (CPF)

To analyze point source impacts from various wind directions, the conditional probability function (CPF) [25] was calculated using source contribution estimates from PMF, 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 hour of a given day to match to the hourly wind data.
Specifically, the CPF is defined as

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

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 in the directions that have high conditional probability values.

3 Results and Discussion

3.1 Source apportionment by PMF modeling

3.1.1 Rajshahi CAMS site:

From the data set, PMF modeling resolved 7 sources for the coarse fraction PM samples. The identified source profiles and the mass contribution of each source for this fraction are presented in Figures 1 and 2, respectively. Figure 3 represent the directional pattern of each source for coarse particle. The first source has the characteristics of Na, Mg, Al, S, Cl, K, Ti, Mn, Cu, Zn and represents Brick kiln. The contribution of this source is from both south-east and north-west direction. The coal that is used in the kiln process contains 4 to 6% sulfur. Due to brick production technology, bricks are produced during dry periods mainly starting from November/December to March/April every year. This profile has seasonal variation and has high contribution in winter [4-5]. The second source profile has the characteristics of Na, Mg, Al, Si, S, Cl, K, Ca, Ti and Fe and represents road dust source and has high contribution in winter mainly from north-west direction. The third source has characteristics of high Na, S, Ca, Fe, Ni, Cu, Zn, Pb and trace amount of Se, Br which is mixed with soil dust profile represents Metal Smelter source and has seasonal variation. This factor is highly influenced by southerly wind.

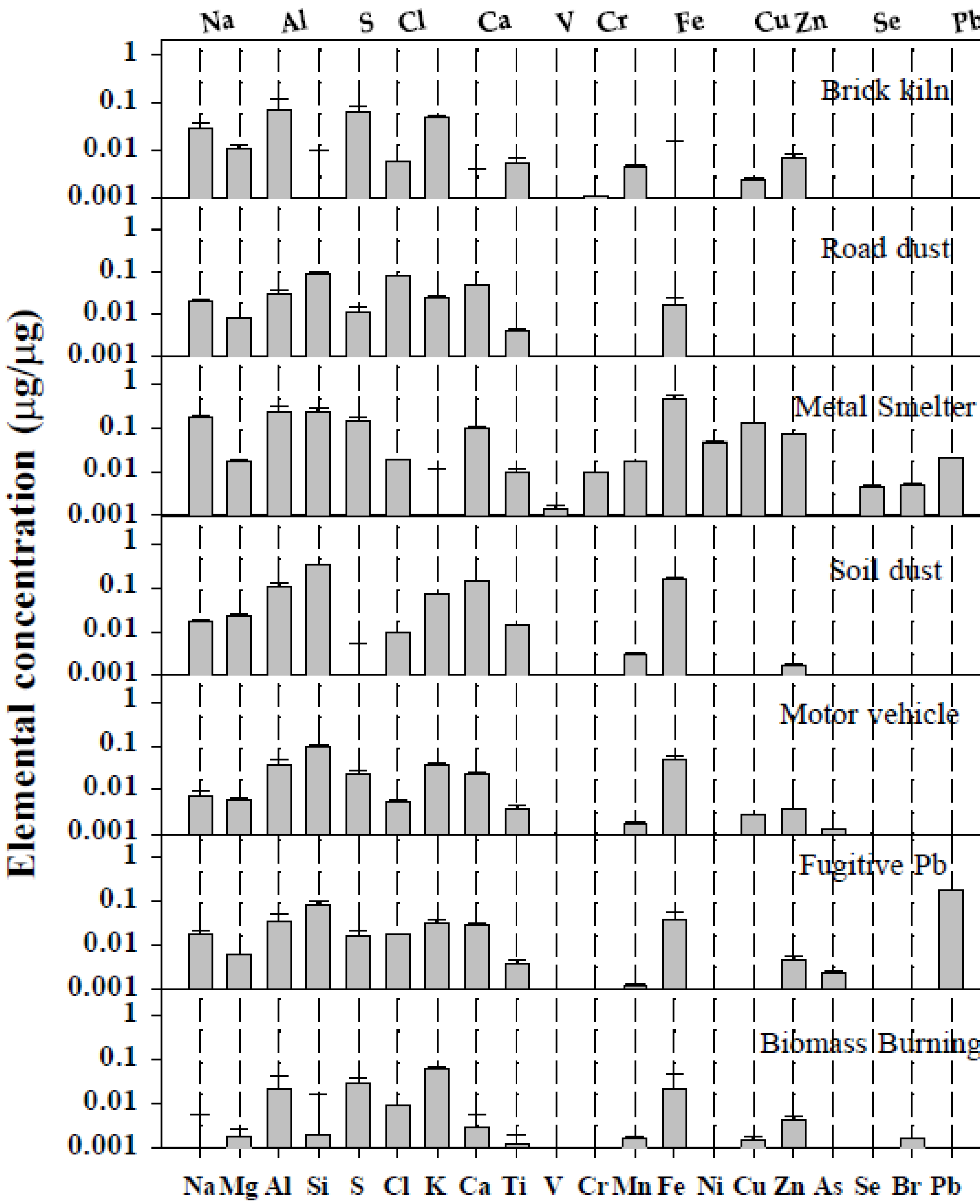

Figure 1: Rajshahi coarse PM source results (source profiles)

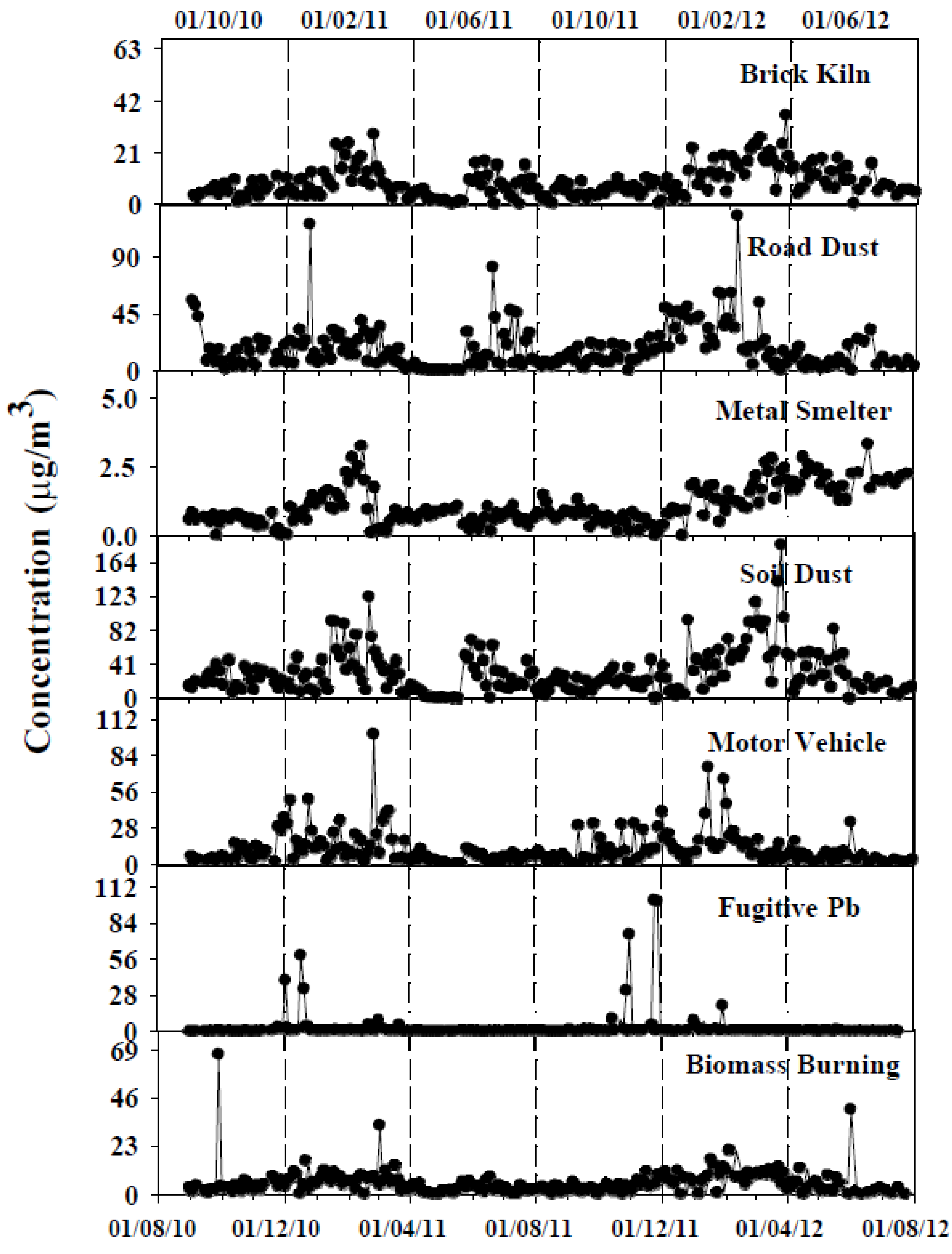

Figure 2: Rajshahi coarse PM time series of source factor contributions.

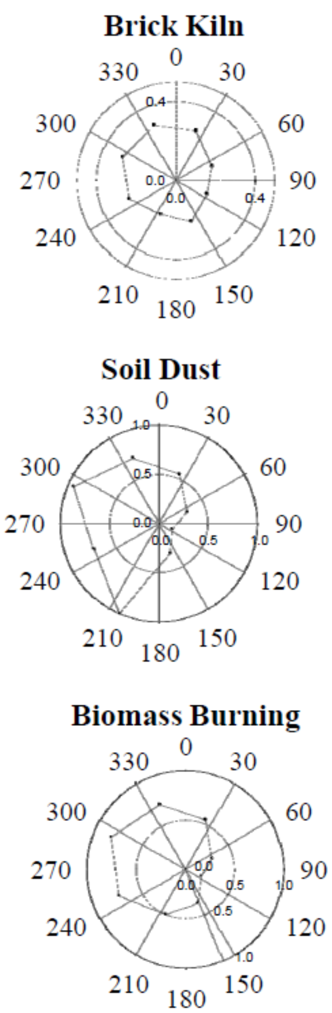

Figure 3: Rajshahi coarse PM direction pattern (in degree) for each source factor indicated (CPF)

The fourth source profile indicated in Figure 1 has the characteristics of high Na, Mg, Al, Si, Cl, K, Ca, Ti, Mn and Fe and represents soil dust and was also influenced by westerly and southerly wind. This profile has seasonal variation and has high contribution in winter. The fifth source has characteristics of S which is mixed with soil dust profile represents motor vehicle source and has seasonal variation that is influenced by westerly winds. The diesel fuel in Bangladesh contains about 3000 ppm sulfur. Heavy duty vehicles mostly use this fuel. However, heavy-duty diesel trucks can only be used in Dhaka from 10 PM to 6 AM in order to reduce their influence on air quality. The sixth source profile has characteristics of high Pb mixed with soil dust particle and represents fugitive Pb source and shows no seasonal variation with few high peaks. The coarse Pb comes from battery reclamation. Breaking up the batteries produces large particles where re-smelting the Pb produces small particle Pb. The factor shows high contribution from south and westerly sectors. The seventh source has characteristics of S, K, Cl, and Fe and trace amount of Mg, Si, Ca, Mn, Cu and Zn that are road dust components along with biomass burning. It has a seasonal variation. The contribution of this factor is influenced by north-westerly wind. It has been found that the coarse fraction carries about 62.2% of soil dust including road dust, as seen in Table 4 (page 34) and explained in the next section.

From the fine PM data from Rajshahi, PMF modeling resolved six source factors (Figures 4 and 5) and the characteristics of source factors are same as in coarse PM. The seasonal influence and the directional pattern (Figure 6) are same as in coarse particles. The fine fraction carries about 85.4% of anthropogenic source contributions such as brick kiln, motor vehicle and wood burning sources, as seen in Table 4 and explained in next section. The regression slope and coefficient for both coarse and fine fractions are given in Table 3. It has been found that directional patterns of fine sources are same as in coarse. The seasonal wind directional pattern for Rajshahi city is shown in Figure 7.

From Figure 5, it has been found that the contribution of high fugitive Pb (172 $\mu g/m^3$ source, biomass burning (404 $\mu g/m^3$) and fine soil dust (164 $\mu g/m^3$) are found on 3 December 2010, 13 February 2011 and 18 March 2012, respectively. From back trajectory calculations (Figure 8,) it is observed that due to the contribution of long-range transport on those days, the overall pollution concentration may have increased.

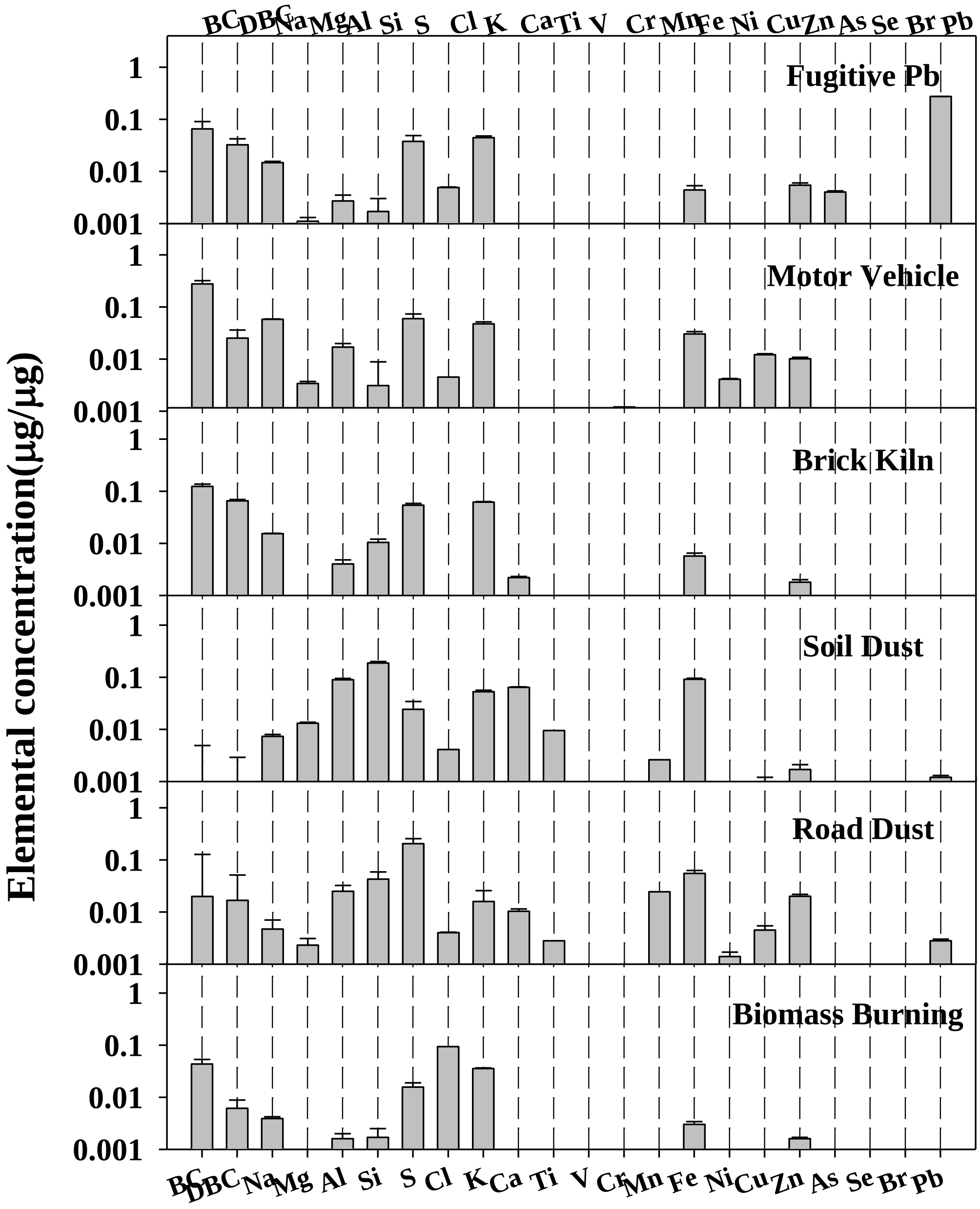

Figure 4: Rajshahi fine PM source profile results

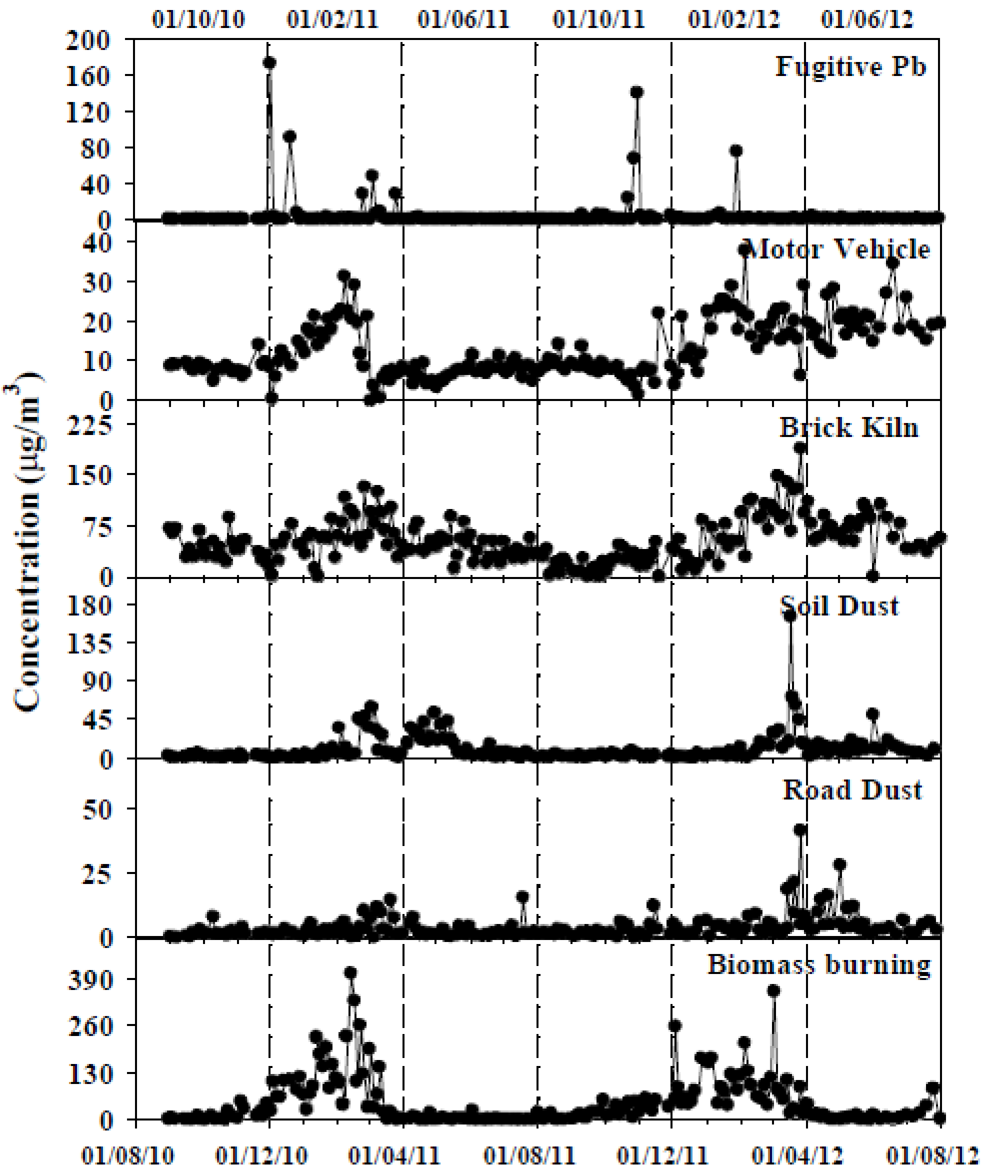

Figure 5: Rajshahi fine PM time series of source factor contributions.

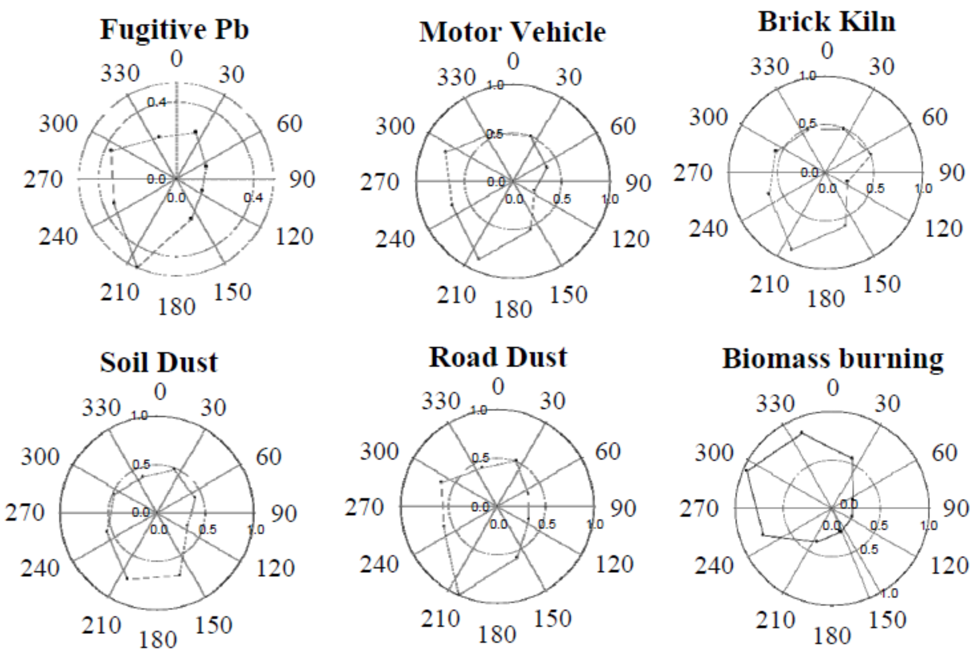

Figure 6: Rajshahi fine PM direction pattern (in degree) for each source factor indicated (CPF)

Table 3: Summary of least squares fit from PMF modelling to the measured mass at each site

Site	**Coarse**		**Fine**	
	Slope	***Coefficient***	***Slope***	***Coefficient***
Rajshahi	0.83	0.78	0.84	0.71
Dhaka	0.81	0.82	0.76	0.69
Khulna	0.62	0.88	0.68	0.66
Chittagong	0.89	0.86	0.84	0.82

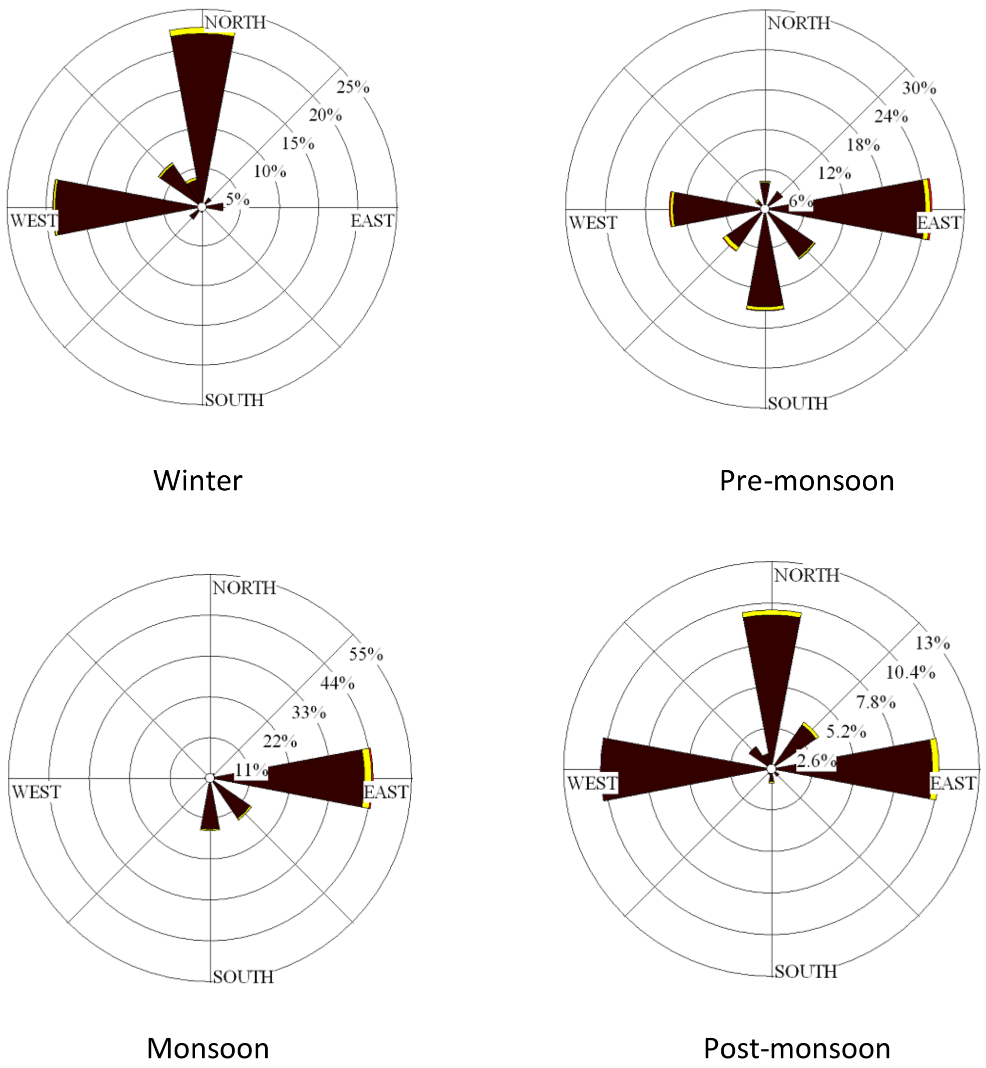

Figure 7: Wind direction pattern (wind roses) for Rajshahi City for four seasons.

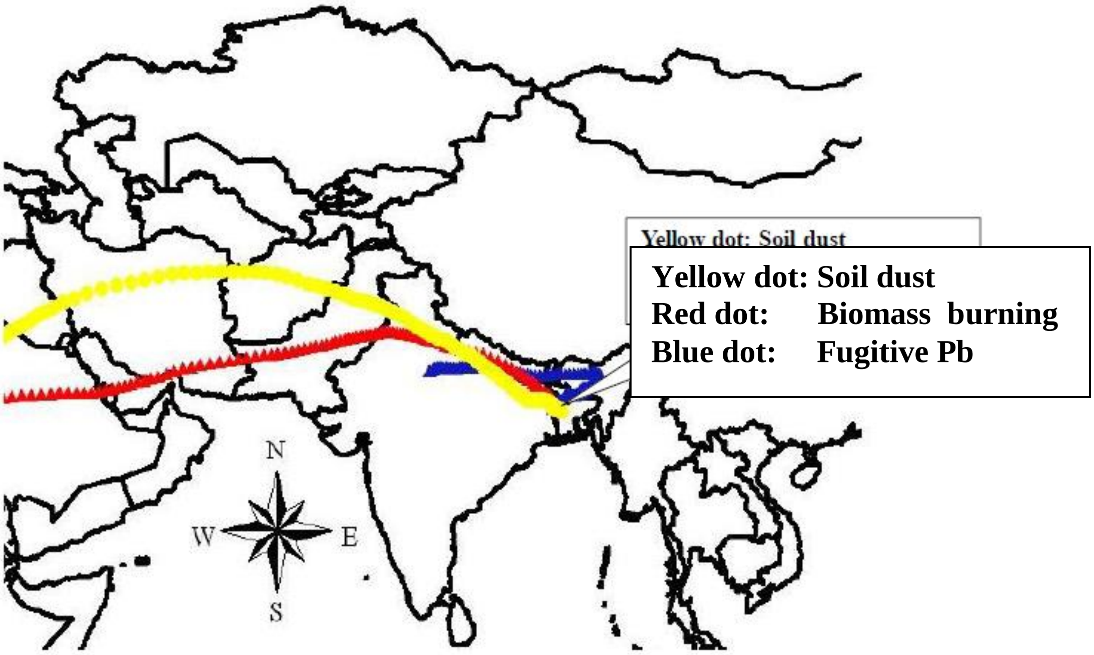

Figure 8: Back trajectory calculations of long-range transport at Rajshahi city for 3. Dec 2010, 13 Feb. 2011 and 18. March 2012.

3.1.2 Dhaka CAMS2 site:

From the Dhaka data, PMF modelling resolved 7 source factors for the coarse fraction PM samples. The identified source factors are brick kilns, road dust, motor vehicles, soil dust, sea salt, Zn source and fugitive Pb. The source profiles and the mass contribution of each source are presented in Figures 9 and 10, respectively. The sources have directional influence as shown in Figure 11. The first source factor has the characteristics of high Al, Si, S, Cl, K, Ca, and Fe and represents brick kiln. The coal that is burnt in the kilns contain 4 to 6% sulfur. Because of the brick production technology, bricks are produced during dry periods mainly starting from November to early March every year. This source profile has seasonal variation and has high contribution in winter [6] and also influenced by north-westerly wind.

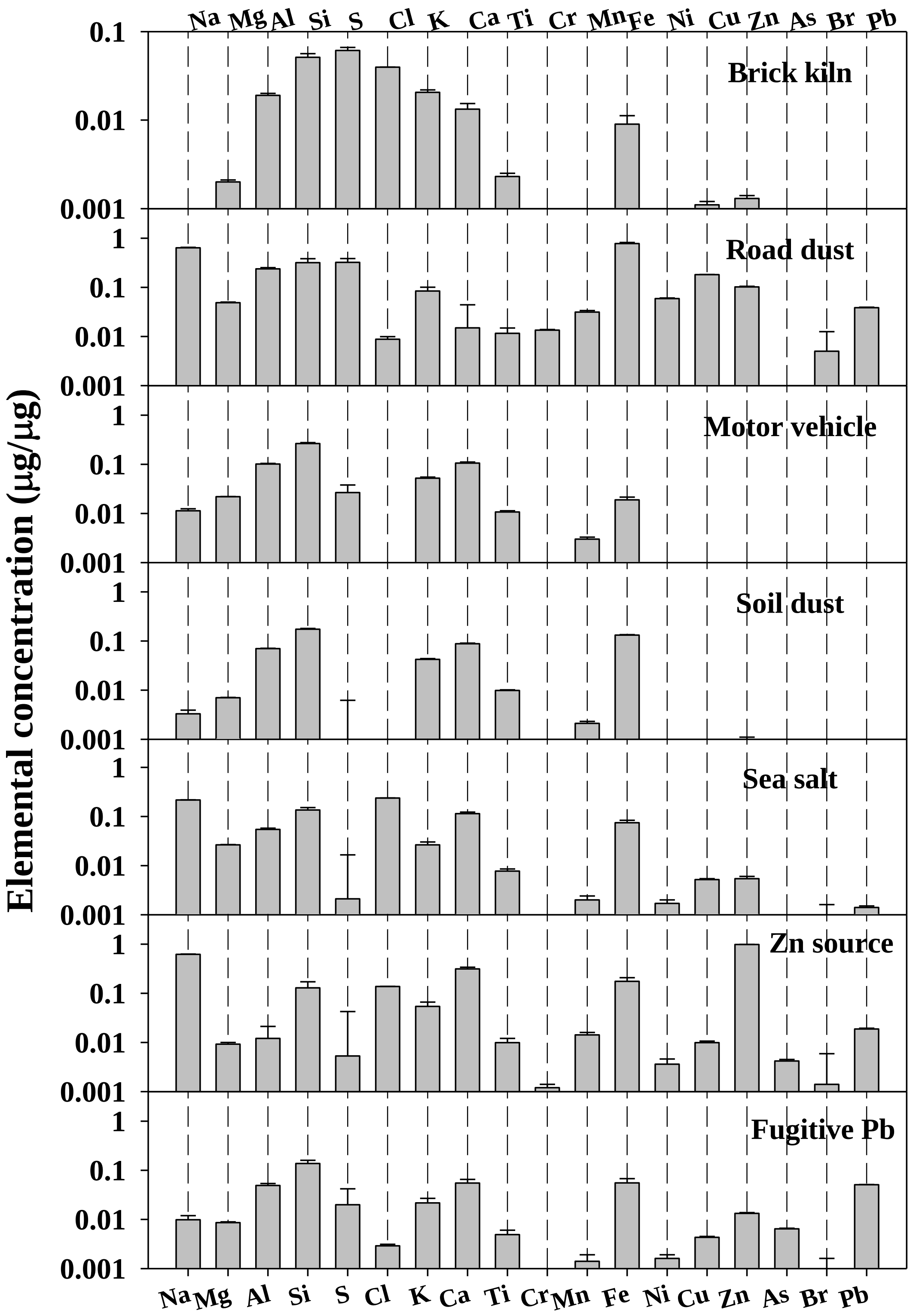

Figure 9: Dhaka coarse PM source profile results

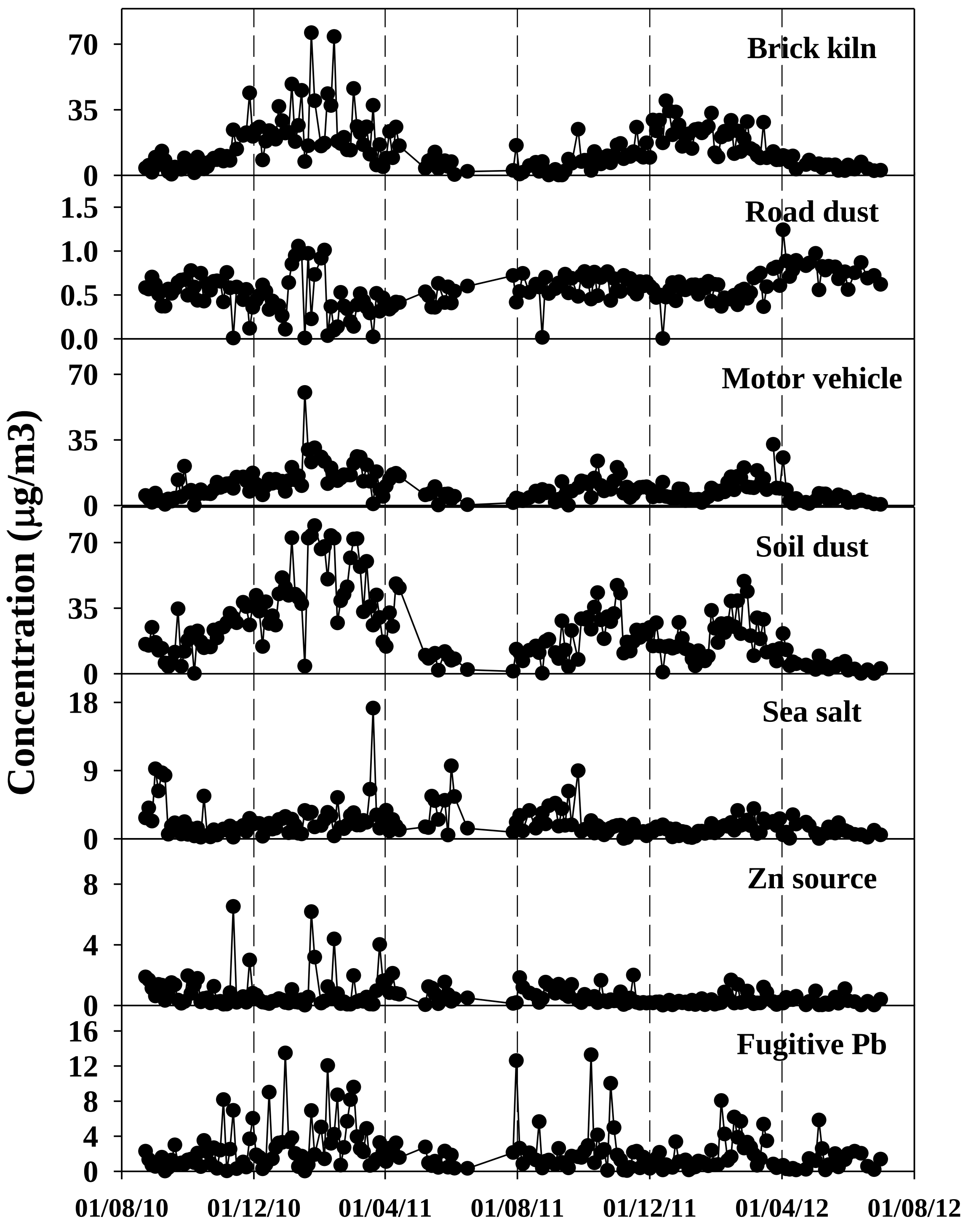

Figure 10: Dhaka coarse PM time series of source factor contributions.

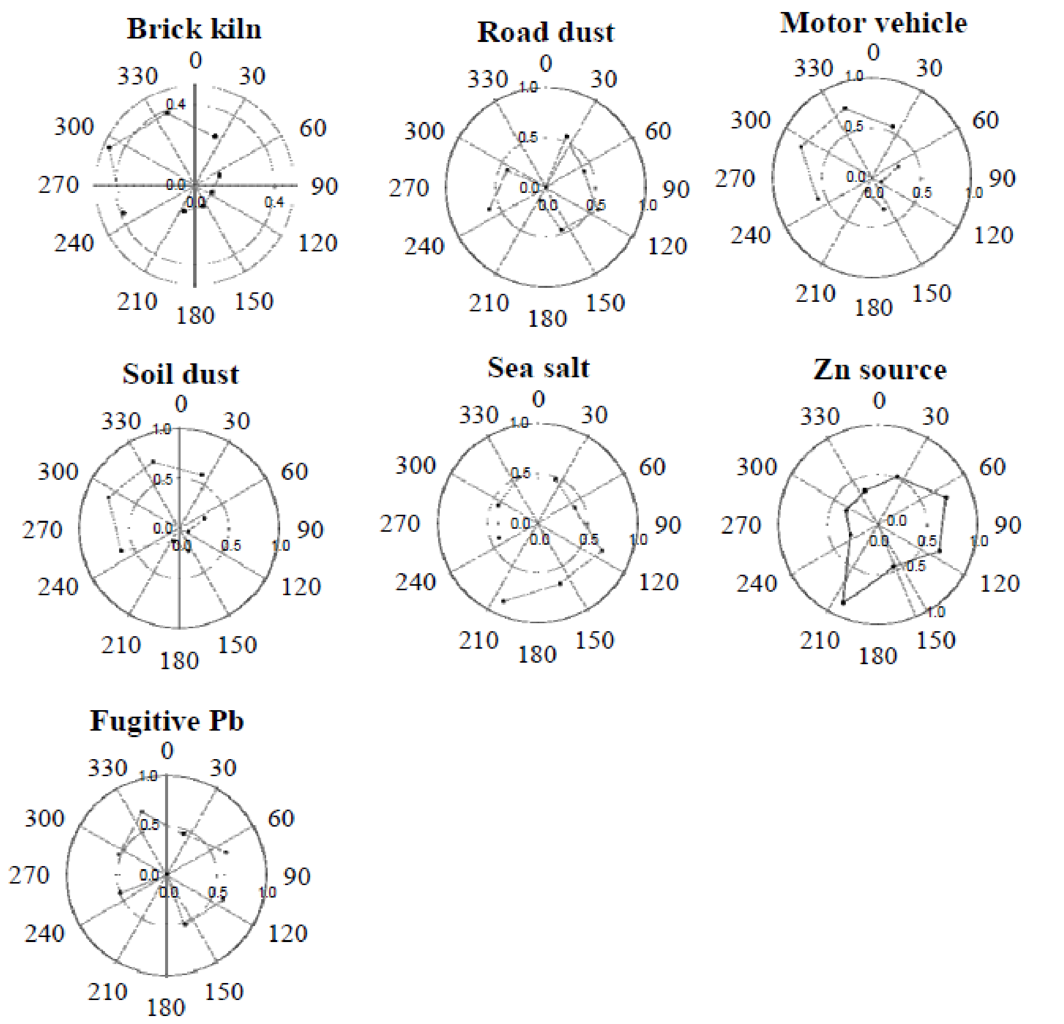

Figure 11: Dhaka coarse PM direction (in degree) pattern for each source factor indicated (CPF)

The second source profile has the characteristics of high BC, Al, Si, S, K, Ti, Mn, Cu, Zn and Pb and represents road dust source and has high contribution in winter season. The third source has S together with road dust and represents motor vehicle source and has seasonal variation. Heavy duty vehicles mostly use high sulfur diesel fuel. The fourth source profile has characteristics of Al, Si, K, Ca, Ti and Fe and represents soil dust source and shows seasonal variation. The fifth source factor has characteristics of Na, and Cl mixed with road dust component and represents sea salt source and has several high peaks during monsoon. The sixth factor has high Na, Cl, Zn, K, Fe, Pb and trace amount of road dust signature and represents a Zn source. Zn may come from the galvanizing factories and to increase the reflectance properties, Pb is added during manufacturing [26]. This factor has several high peaks with no seasonal variation. The seventh source is fugitive Pb factor and has characteristics of high Pb and mixed with Na, Mg, Al, Si, S, Cl, K, Ca, Fe, Zn, and As. This source has no seasonal variation and has several high contributions throughout the year. The main source of Pb in fine PM is from battery recycling.

From fine PM data, PMF modelling resolved seven source factors and the characteristics of the sources are same as for coarse PM (Figure 12). The seasonal variation (Figure 13) and the directional pattern (Figure 14) are similar as for coarse particles. The fine fraction carries about 68.4% of anthropogenic sources such as brick kiln, motor vehicle and wood burning sources, as explained in next section (See also Table 4). The regression slope and coefficient for both coarse and fine fractions are given in Table 3. It has been found that directional patterns of source contributions in the fine PM are same as in coarse fraction results.

The seasonal wind directional pattern for Dhaka city is shown in Figure 15. From Figure 13, it can be seen that the contribution of high fugitive Pb source (21.3 mg/m3), brick kiln (102 mg/m3) , and soil dust (18.9 mg/m3) sources peak on 12 November 2010, 3 March 2011 and 14 April 2011, respectively. From back trajectory analyses it has found that the contributions of long-range transport on those days may have increased the measured pollution concentrations (see Figure 16).

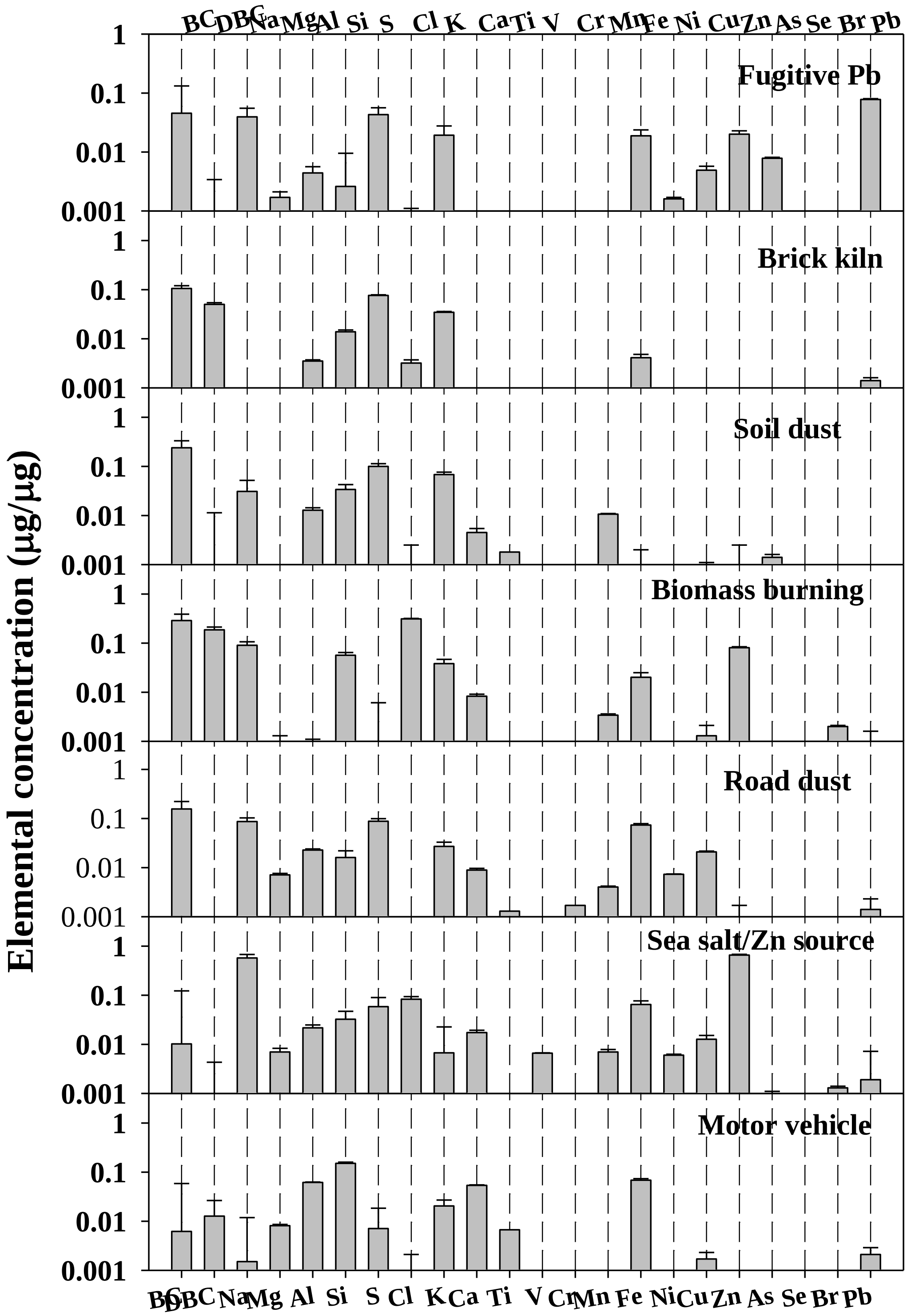

Figure 12: Dhaka fine PM source profile results

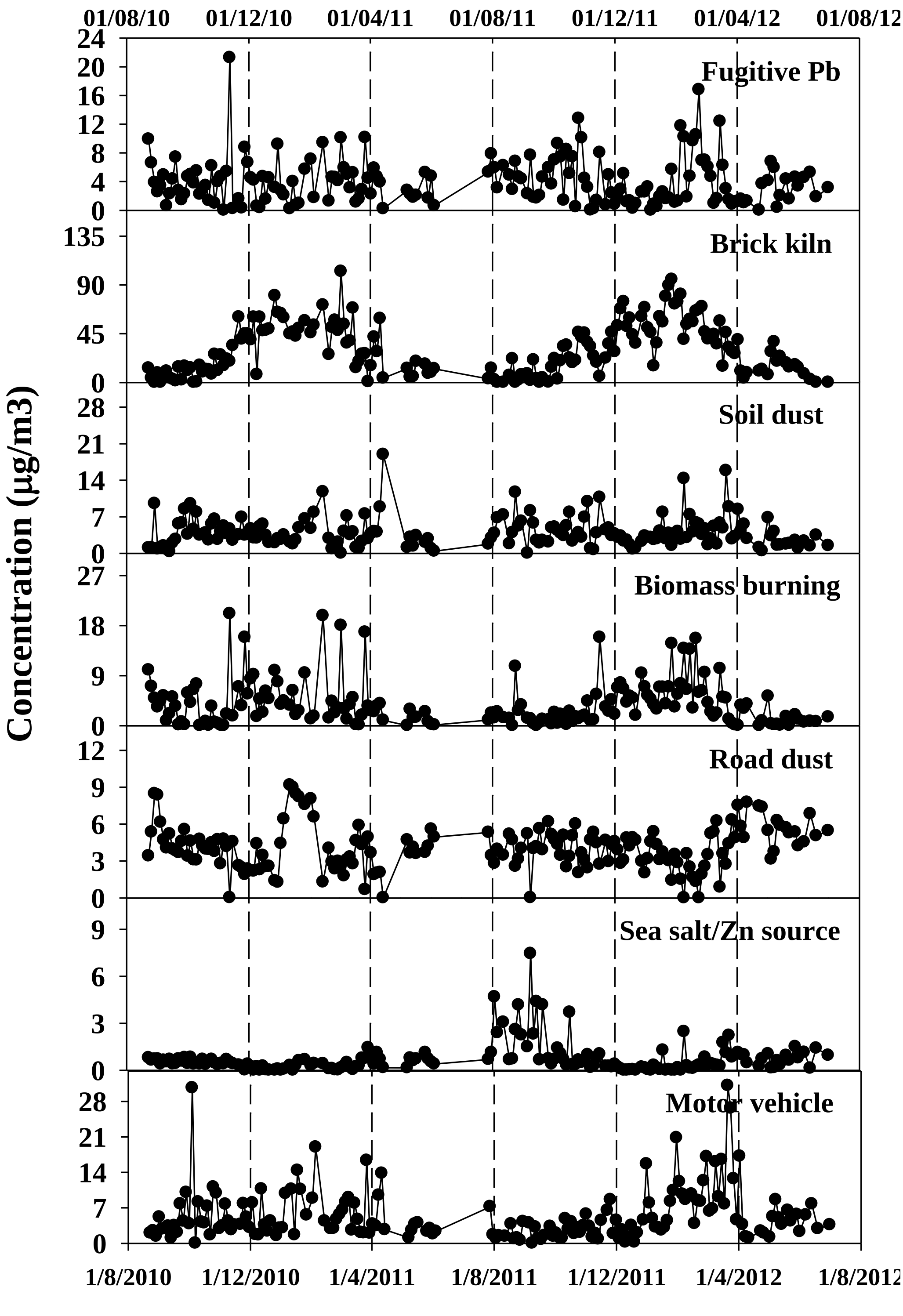

Figure 13: Dhaka fine PM time series of source factor contributions

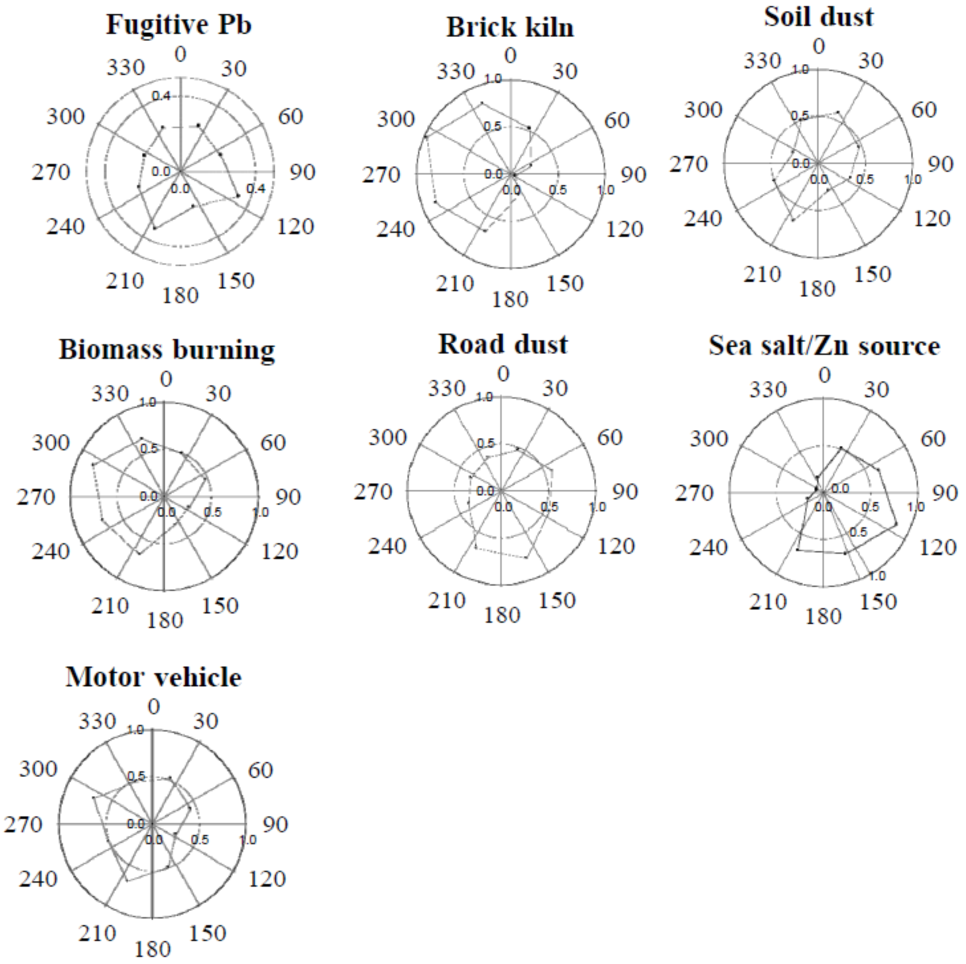

Figure 14: Dhaka pattern for each source factor indicated (CPF) as a function of wind directions (degree)

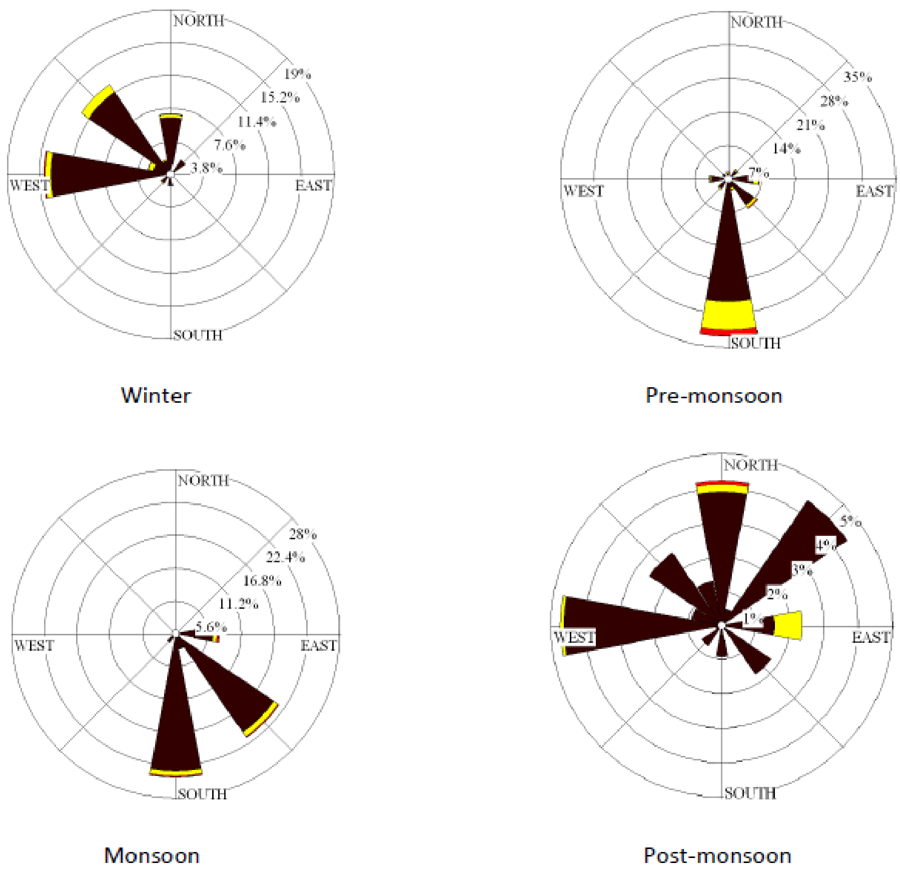

Figure 15: Seasonal wind direction pattern (wind roses) for Dhaka City

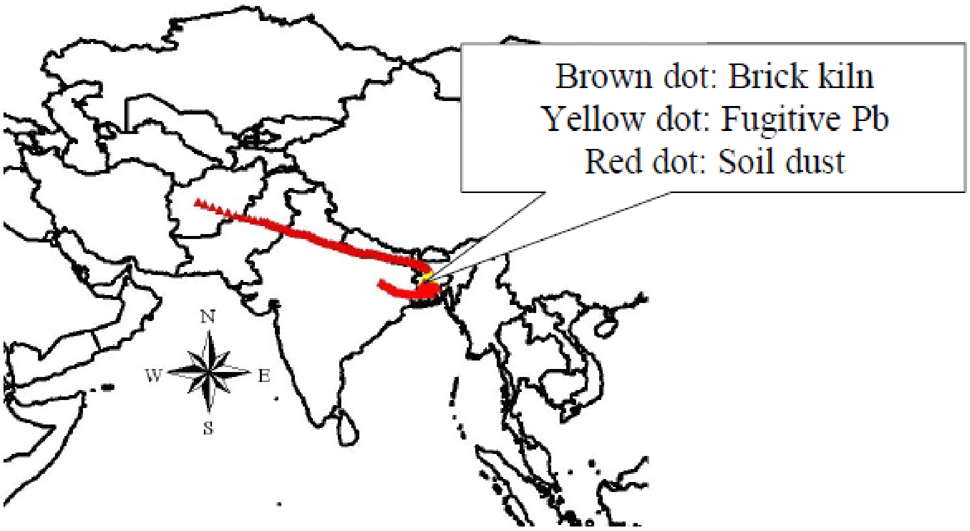

Figure 16: Back trajectory calculations of long-range transport at Dhaka

3.1.3 Khulna CAMS site:

From the data of Khulna CAMS site, PMF modeling resolved 7 source factors for both coarse and fine fractions of PM samples. The identified source factors are brick kiln, wood burning, metal smelters, road dust, motor vehicles, soil dust, sea salt, and fugitive Pb sources depending on the PM fraction.

The source profiles, the mass contribution and CPF plot of each source factor for coarse PM are presented in Figures 17, 18, and 19, respectively. Figure 20, 21 and 22 show the identified source factors, their seasonal variations and the directional contributions for fine PM. The characteristics of sources are similar to those discussed for Rajshahi and Dhaka sites. The regression slopes and coefficients of both coarse and fine fractions are given in Table 3.
Figure 23 shows the wind roses for Khulna city. From Figure 21, it can be seen that the contribution of high brick kiln (113 µg/m^3), motor vehicle (92.0 µg/m^3 and fugitive Pb source (37.0 µg/m^3), sources peak on 23 January 2011, 3 January 2012 and 17 April 2012, respectively. From Figure 24, it has been found that the contributions of long-range transport on those days have increased the measured pollution concentrations. Figure 24 shows the long- range transport of fine sources at Khulna site.

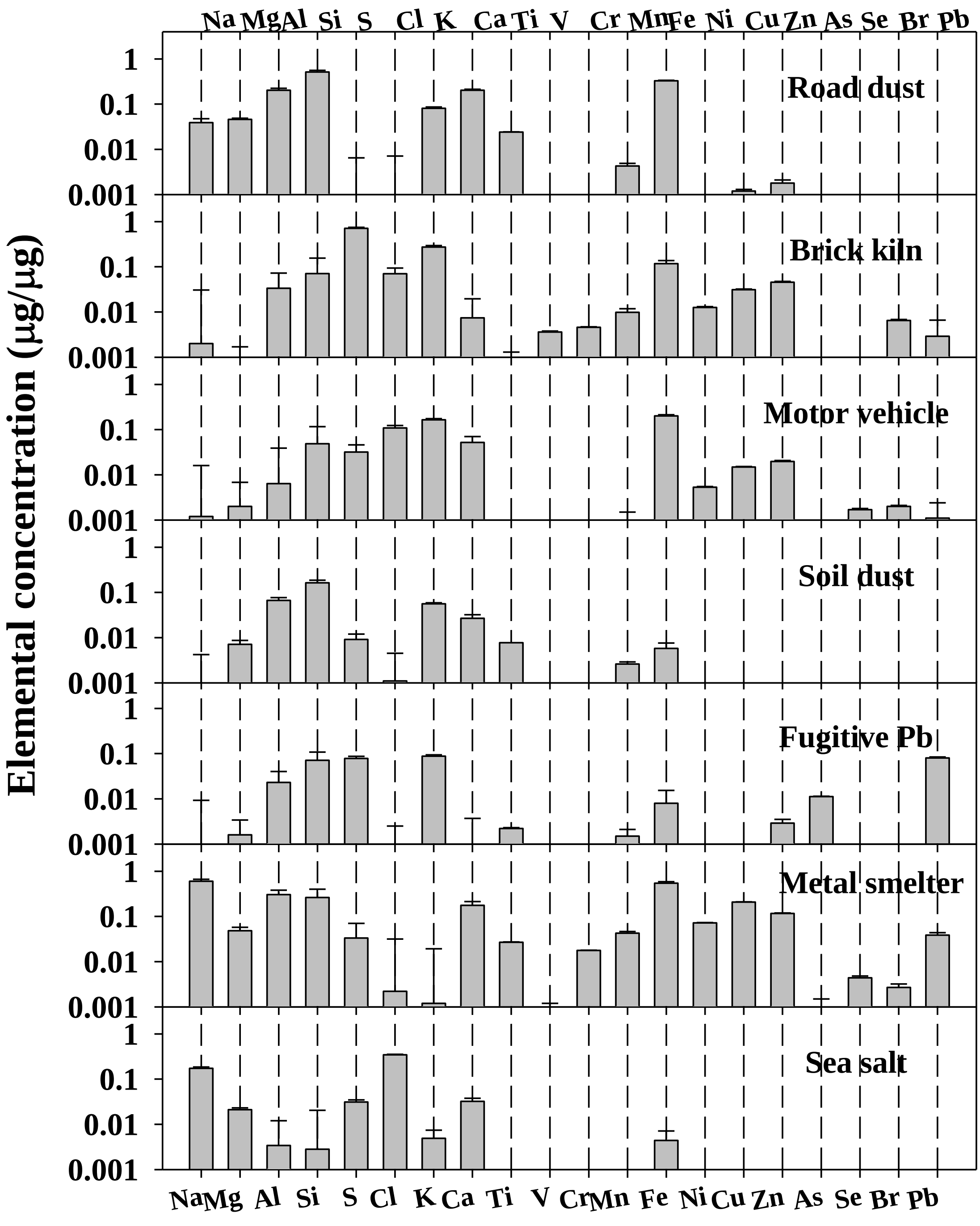

Figure 17: Khulna coarse PM source profile results

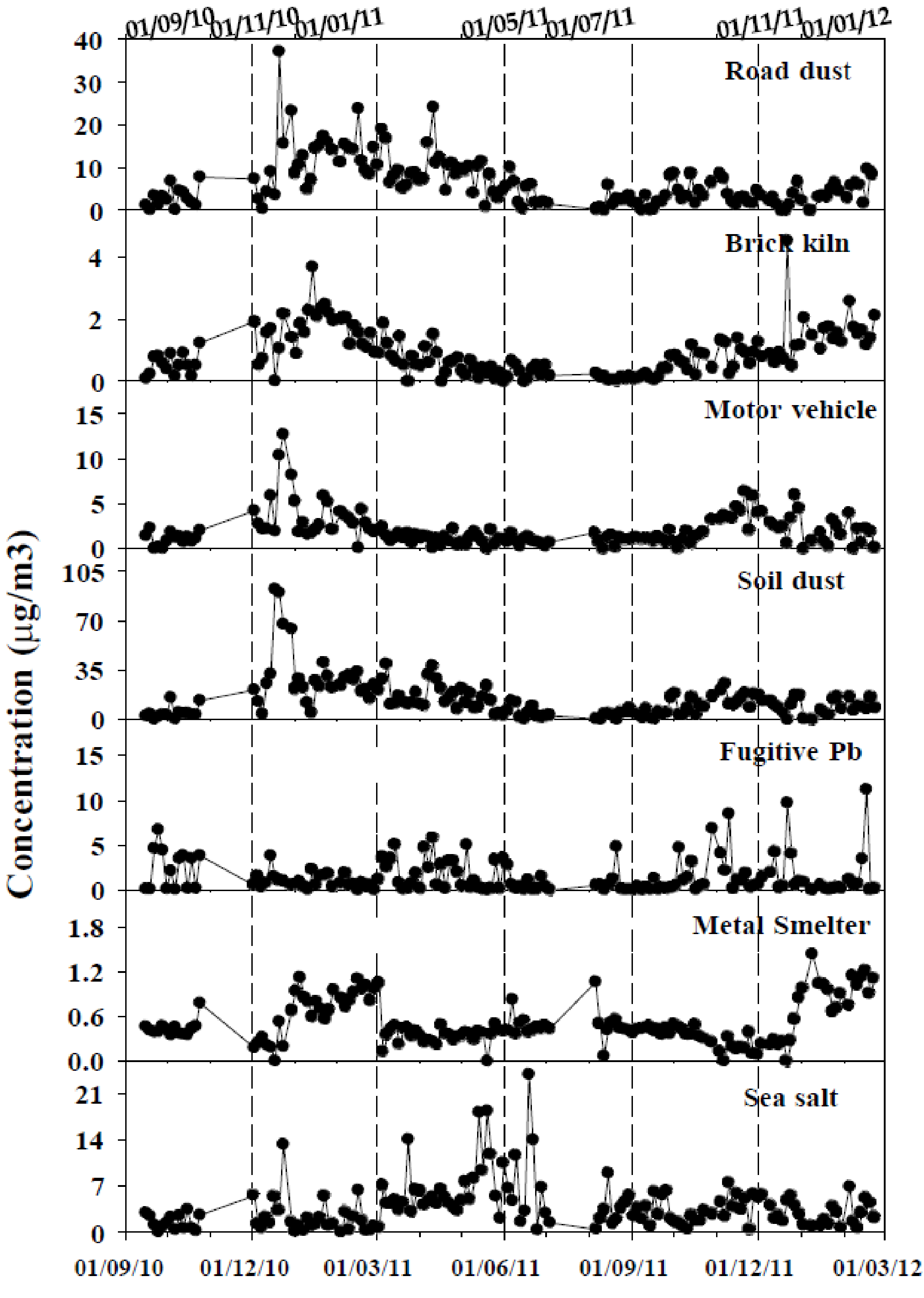

Figure 18: Khulna coarse PM time series of source factor contributions

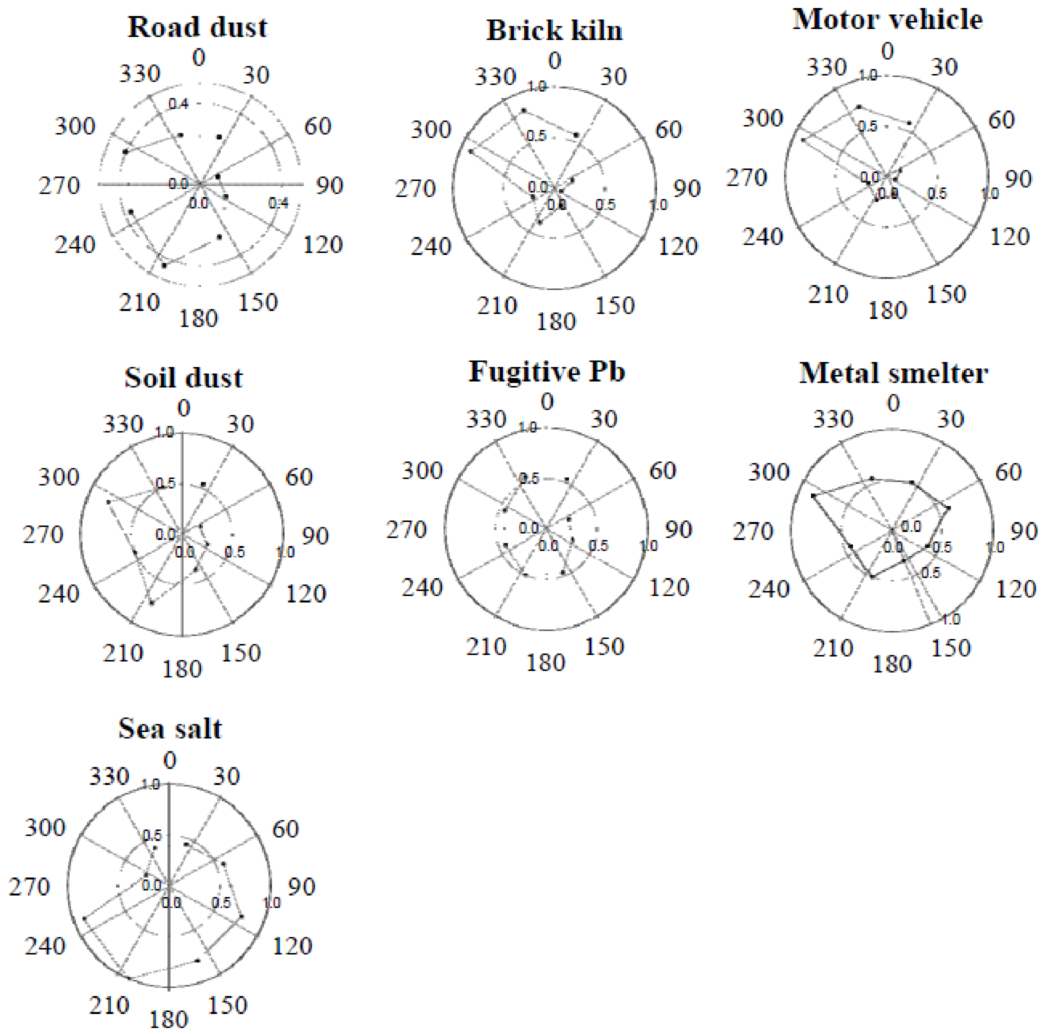

Figure 19: Khulna: fine PM direction pattern for each source factor indicated (CPF) as a function of wind directions (in degrees).

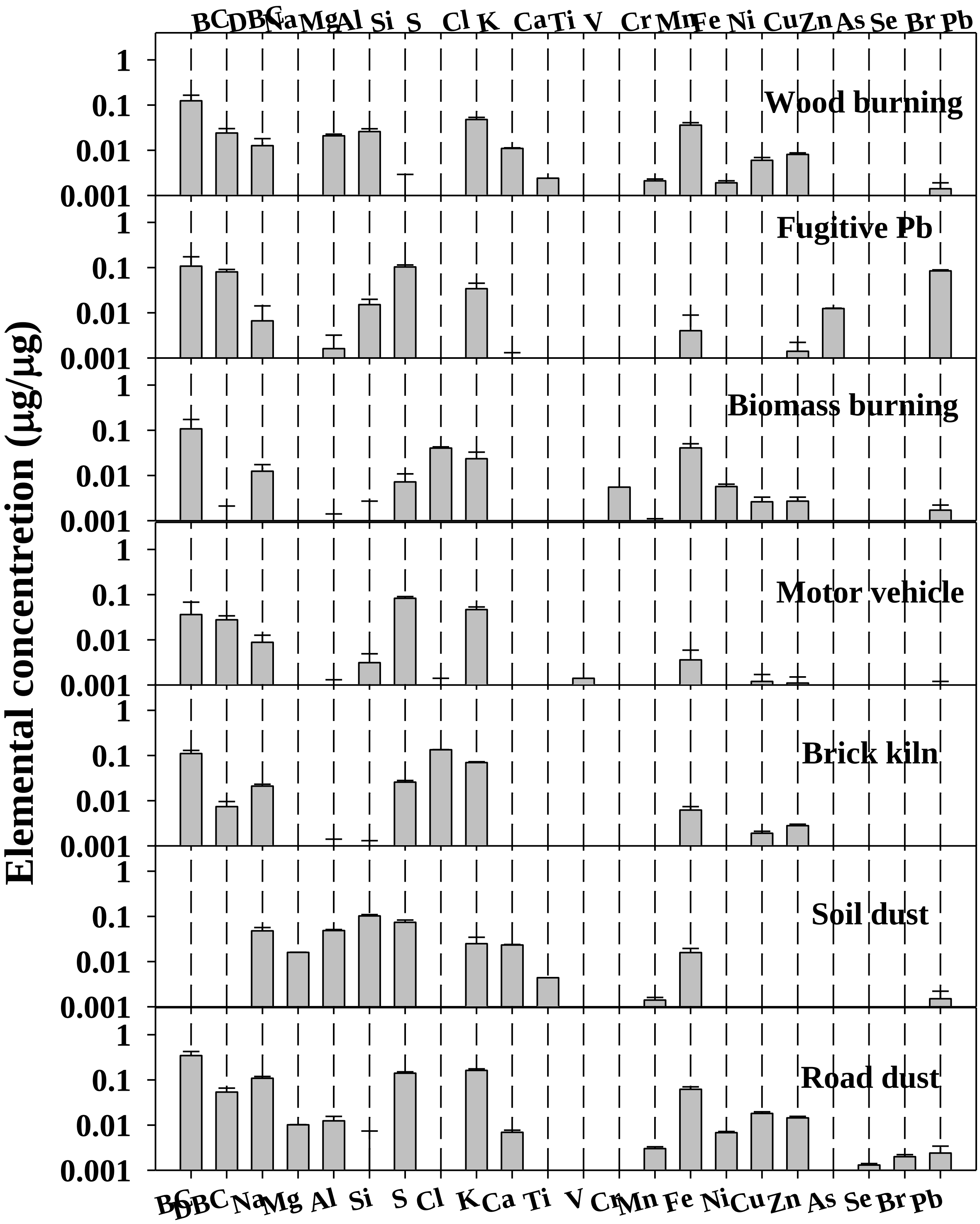

Figure 20: Khulna fine PM source profile results

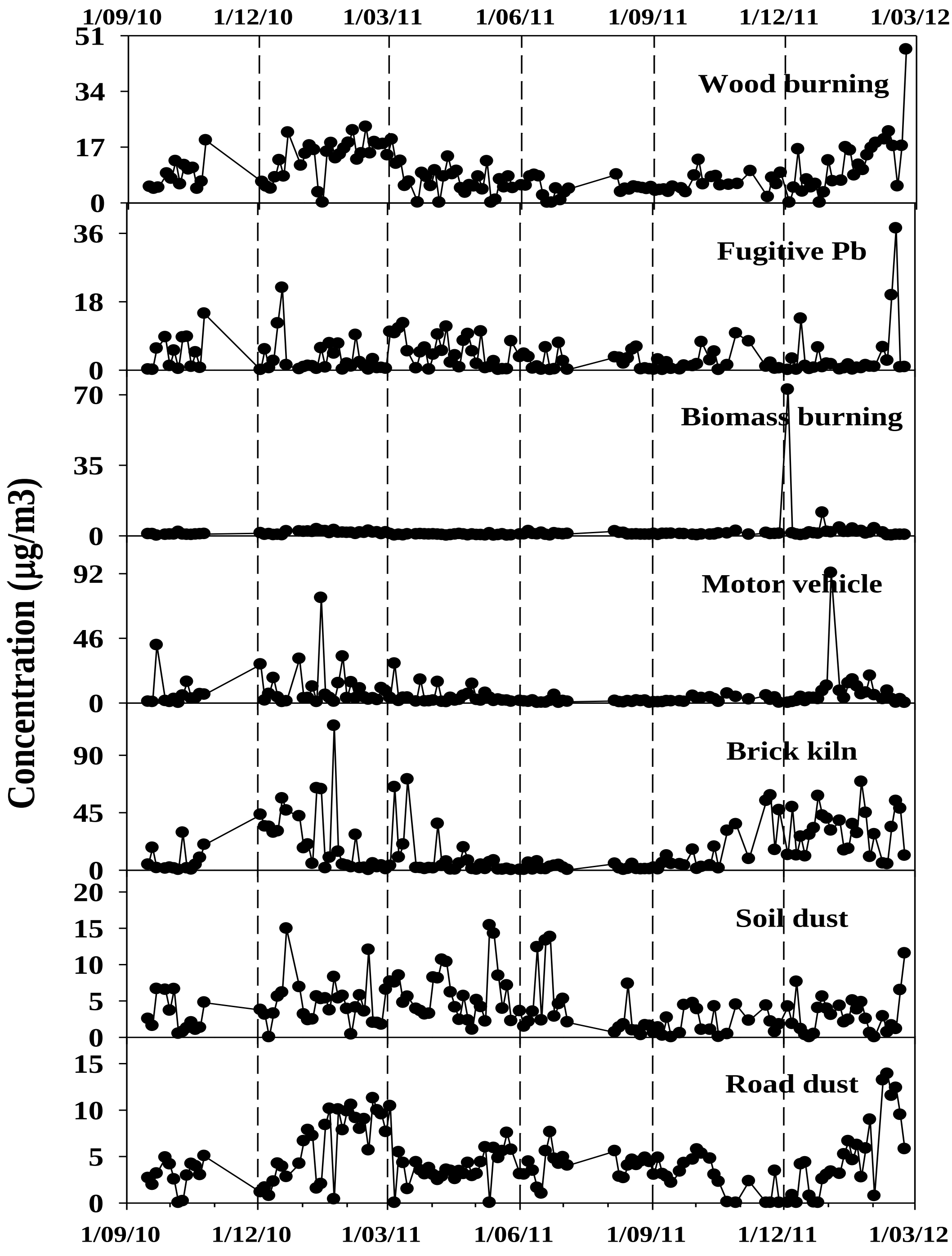

Figure 21: Khulna fine PM time series of source factor contributions

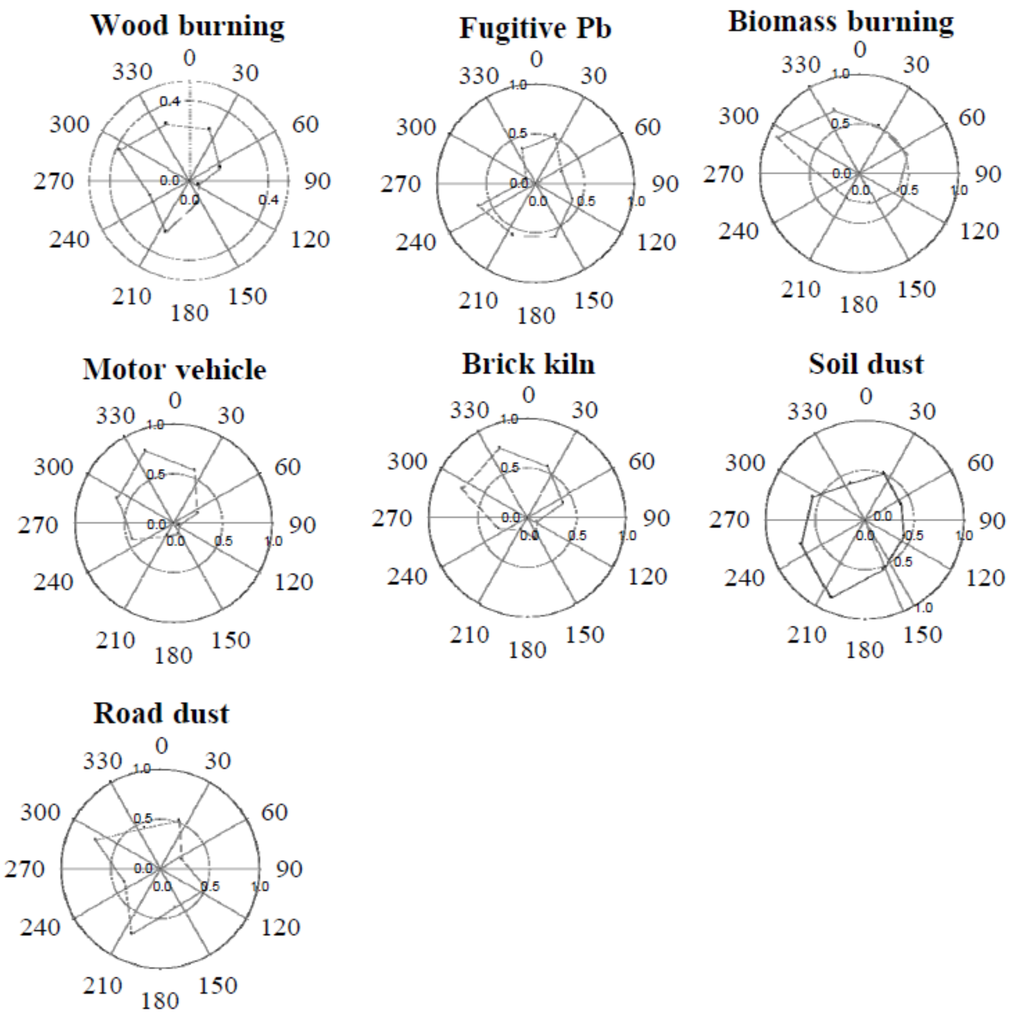

Figure 22: Khulna: Fine PM direction pattern for each source factor indicated (CPF) as a function of wind directions (in degree)

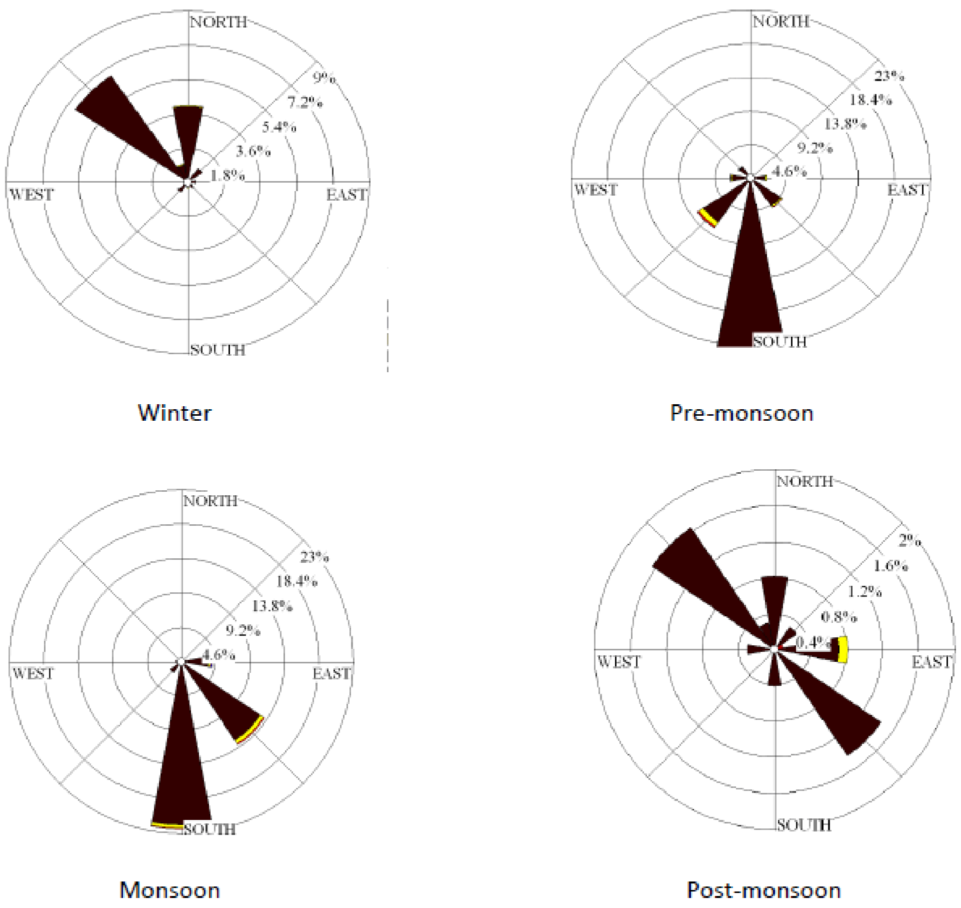

Figure 23: Seasonal wind direction distributions (wind roses) at Kulna.

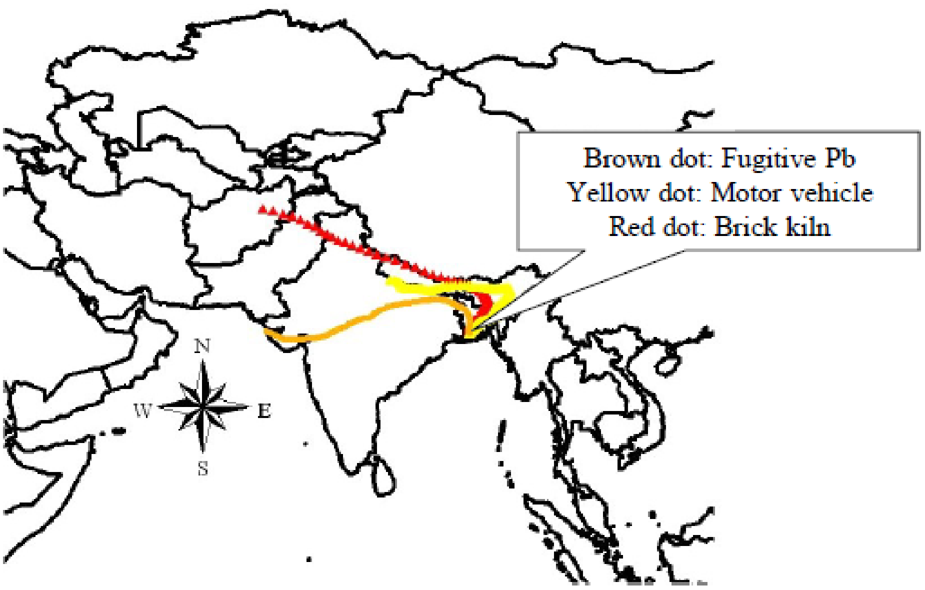

Figure 24: Back trajectory calculations of long-range transport at Khulna city

3.1.4 Chittagong CAMS site:

From the data of Chittagong CAMS site, PMF modelling resolved 7 source factors for both coarse and fine PM fractions. The identified sources are brick kilns, wood burning, metal smelters, road dust, motor vehicles, soil dust, sea salt, Zn source and fugitive Pb sources depending on the size fraction of PM.

The source profiles and the mass contribution of each source are presented in Figures 25, 26, 28 and 29, respectively. The source factors contributions have directional influence (Figures 27 and 30). The characteristics of sources are similar to those discussed for the other three sites. The regression slope and coefficient for both coarse and fine fractions are given in Table 3. From Figure 29, it can be seen that the contribution of high wood burning and Sea salt/Zn (97.0 and 99.0 µg/m3), wood burning (68.2. µg/m3) and sea salt/Zn (62.7 µg/m3) and brick kiln source (68.6 µg/m3), sources peak on 04 February 2011, 23 February 2011, 08 February 2012 and 17 February 2012, respectively. Figure 31 shows wind directional patterns for Chittagong. Figure 32 shows the air mass trajectories for some selected days with long-range transport of fine particles at Chittagong site.

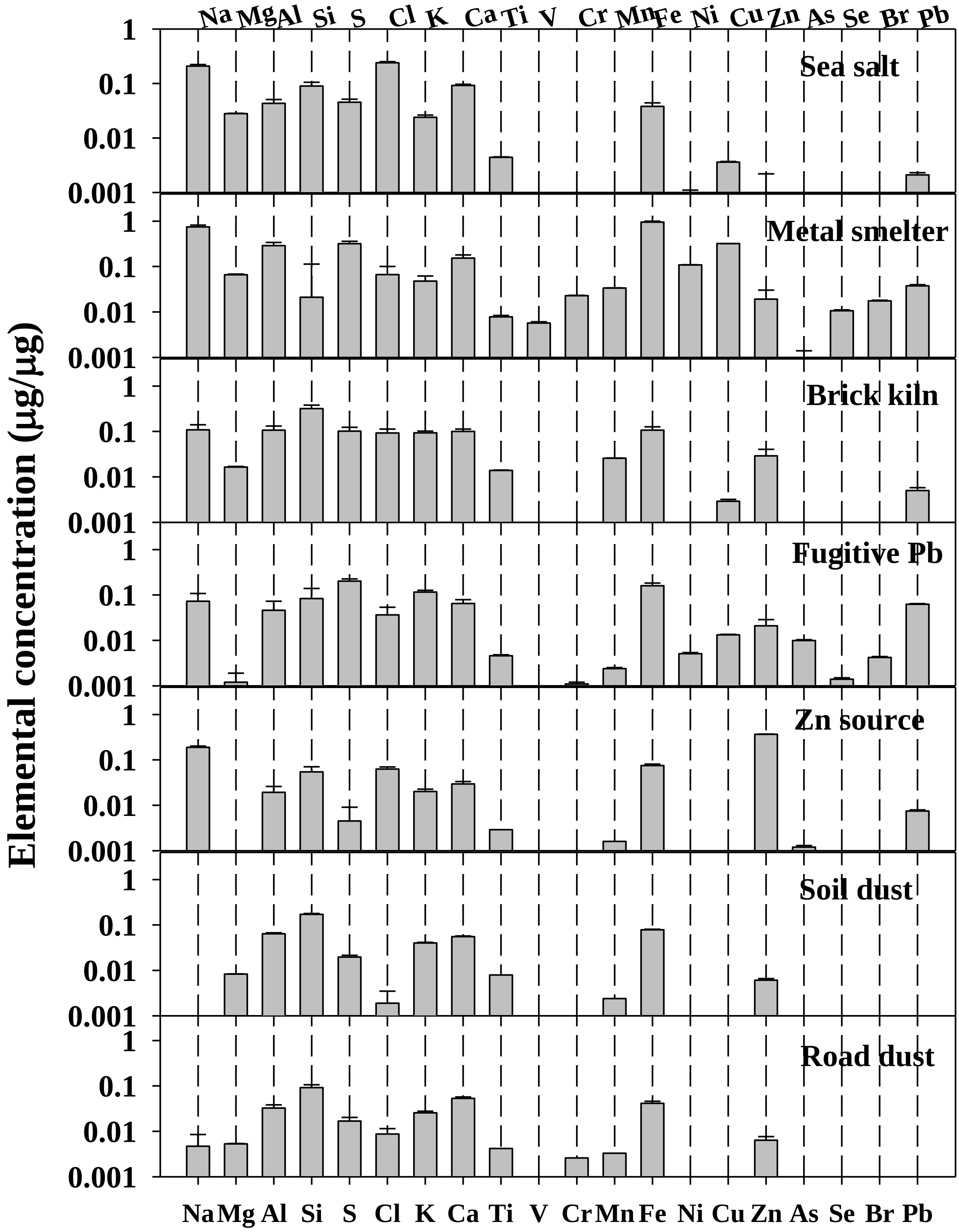

Figure 25: Chittagong coarse PM source results (source profiles).

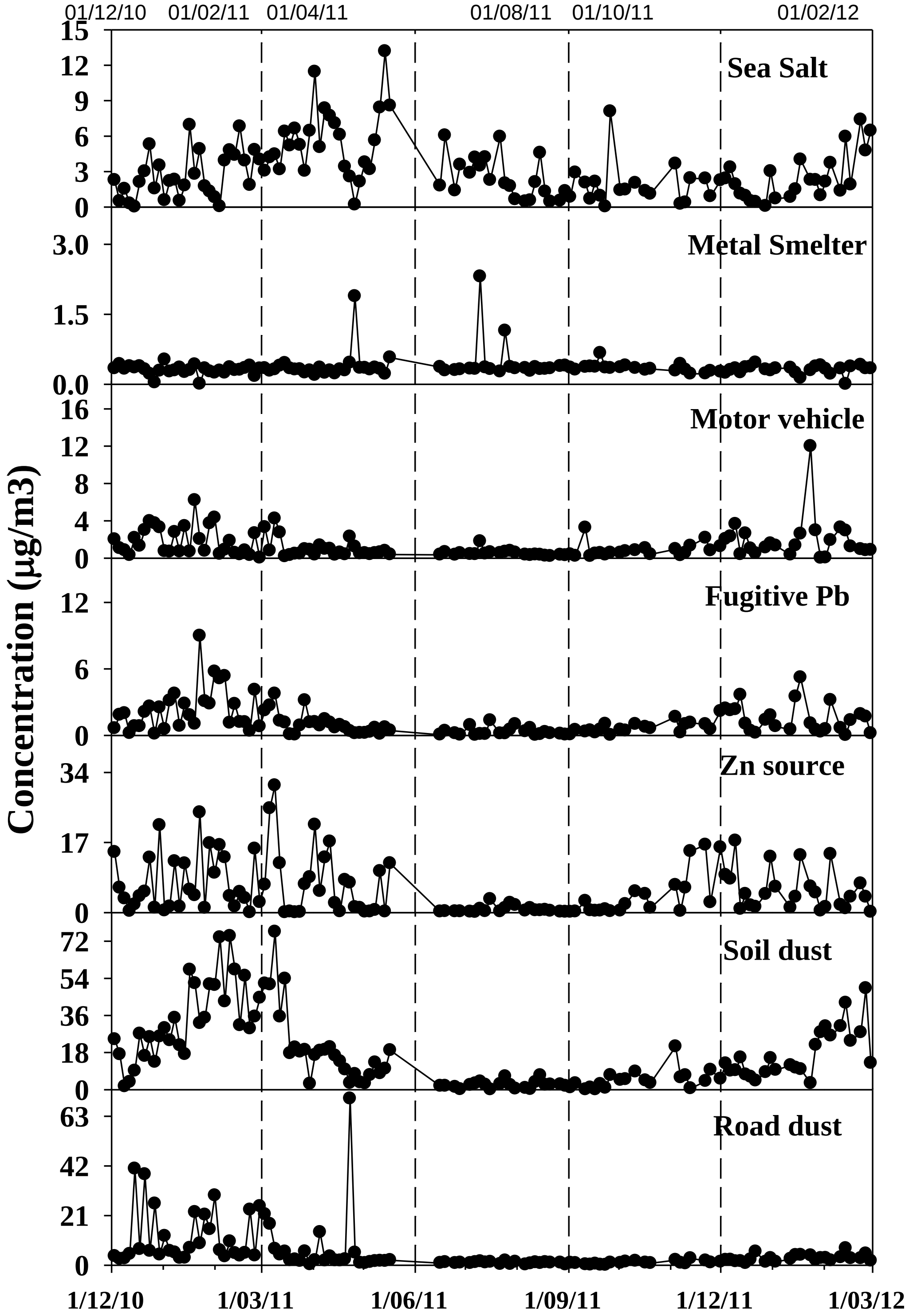

Figure 26: Chittagong coarse PM time series of source factor contributions

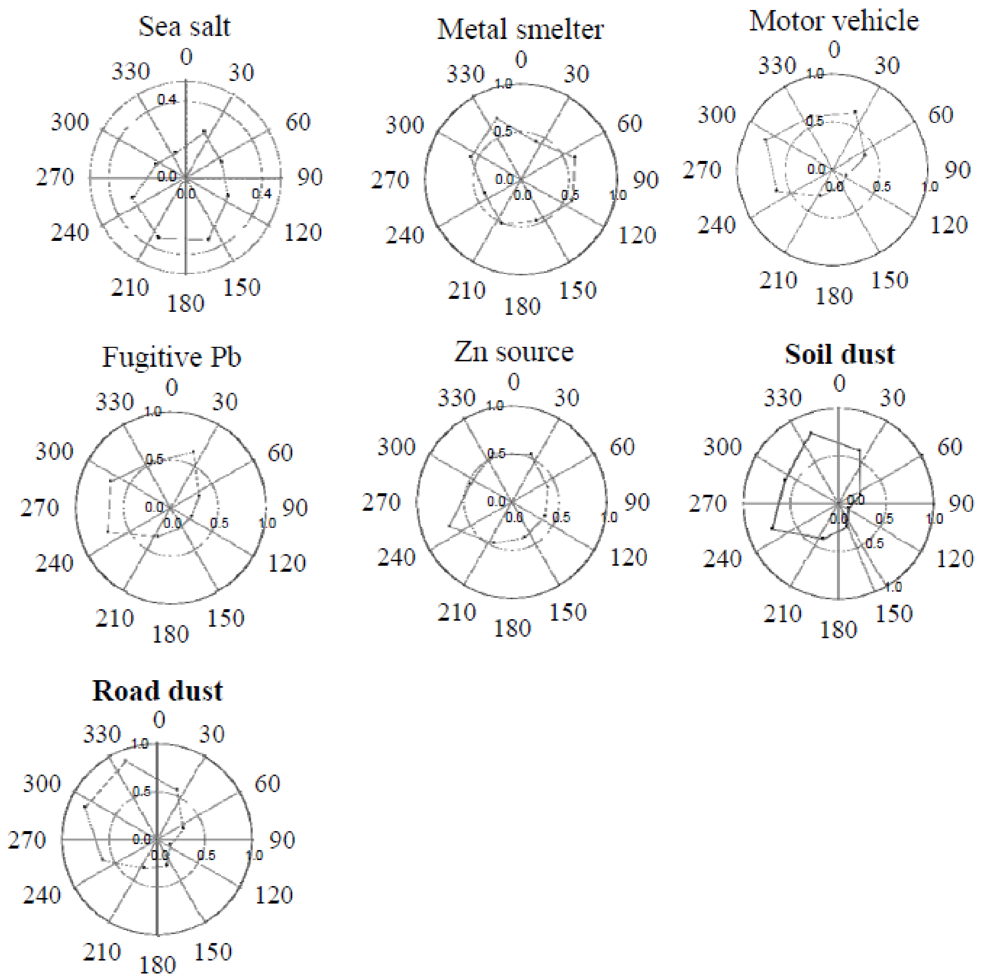

Figure 27: Chittagong coarse PM direction (in degree) pattern for each source factor indicated (CPF)

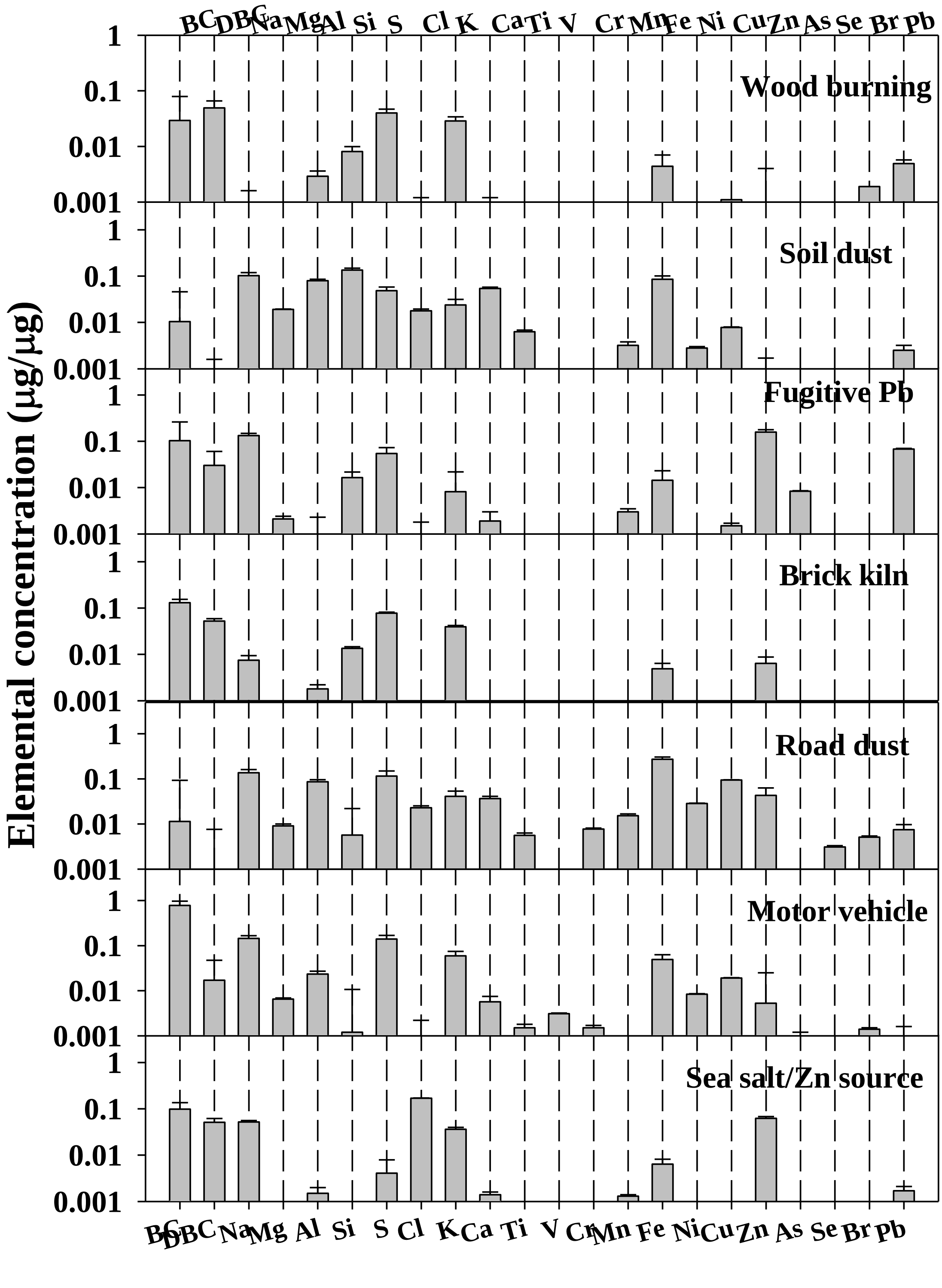

Figure 28: Source profiles for fine PM measured in Chittagong

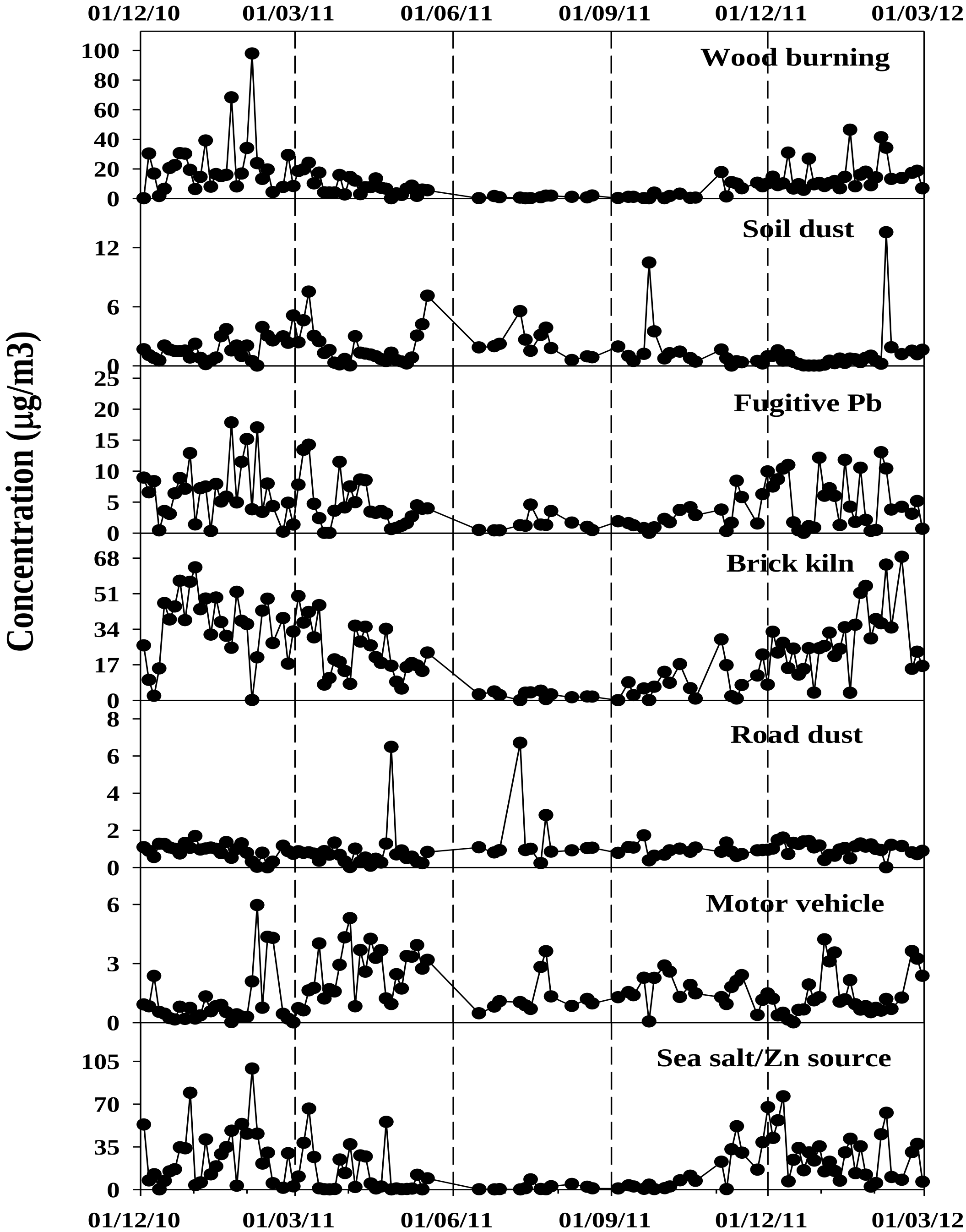

Figure 29: Time series of source factors in fine PM measured in Chittagong

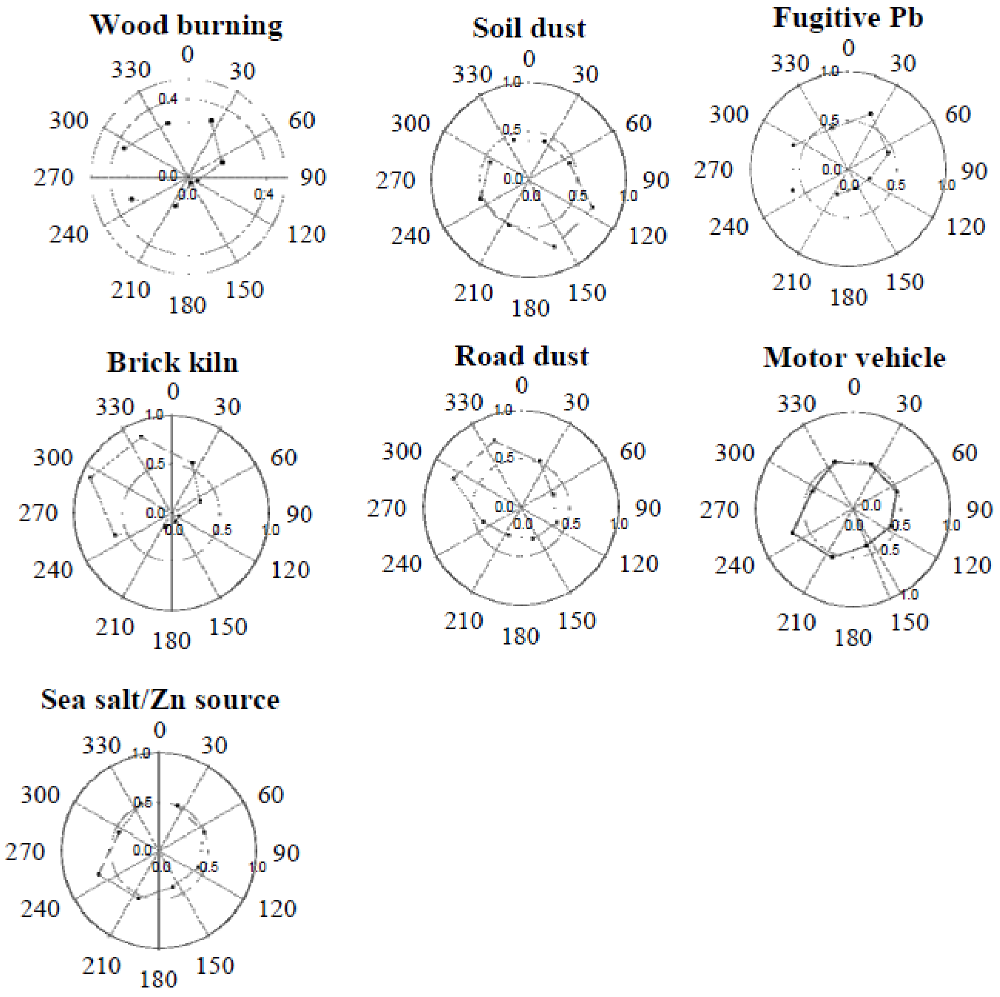

Figure 30: Chittagong: Fine PM direction pattern for each source factor indicated (CPF) as a function of wind directions (degree).

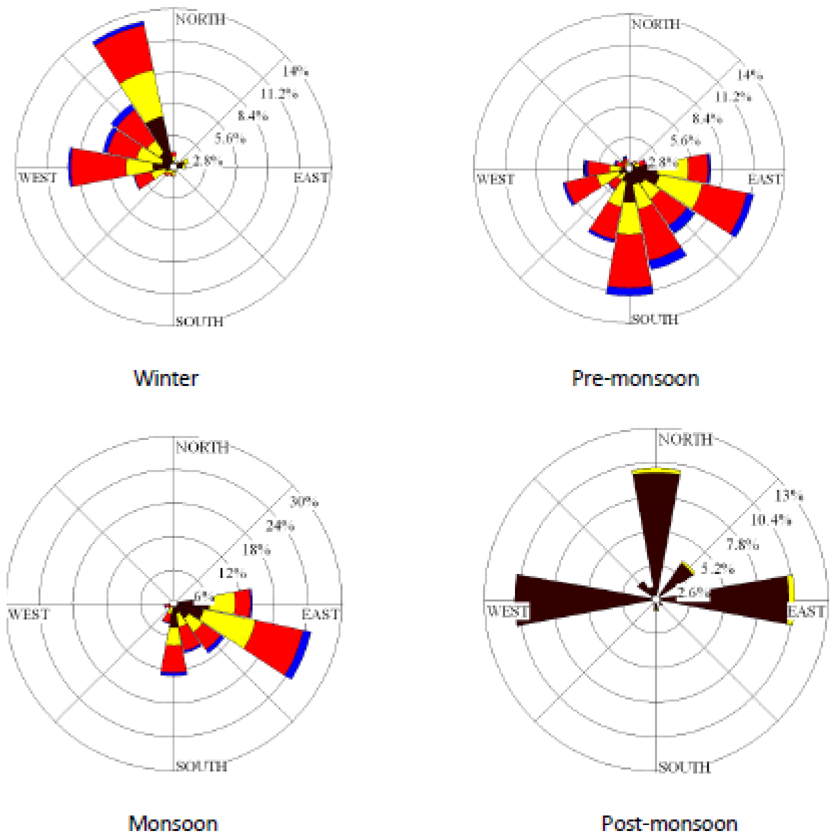

Figure 31: Seasonal wind frequencies (wind roses) for Chittagong.

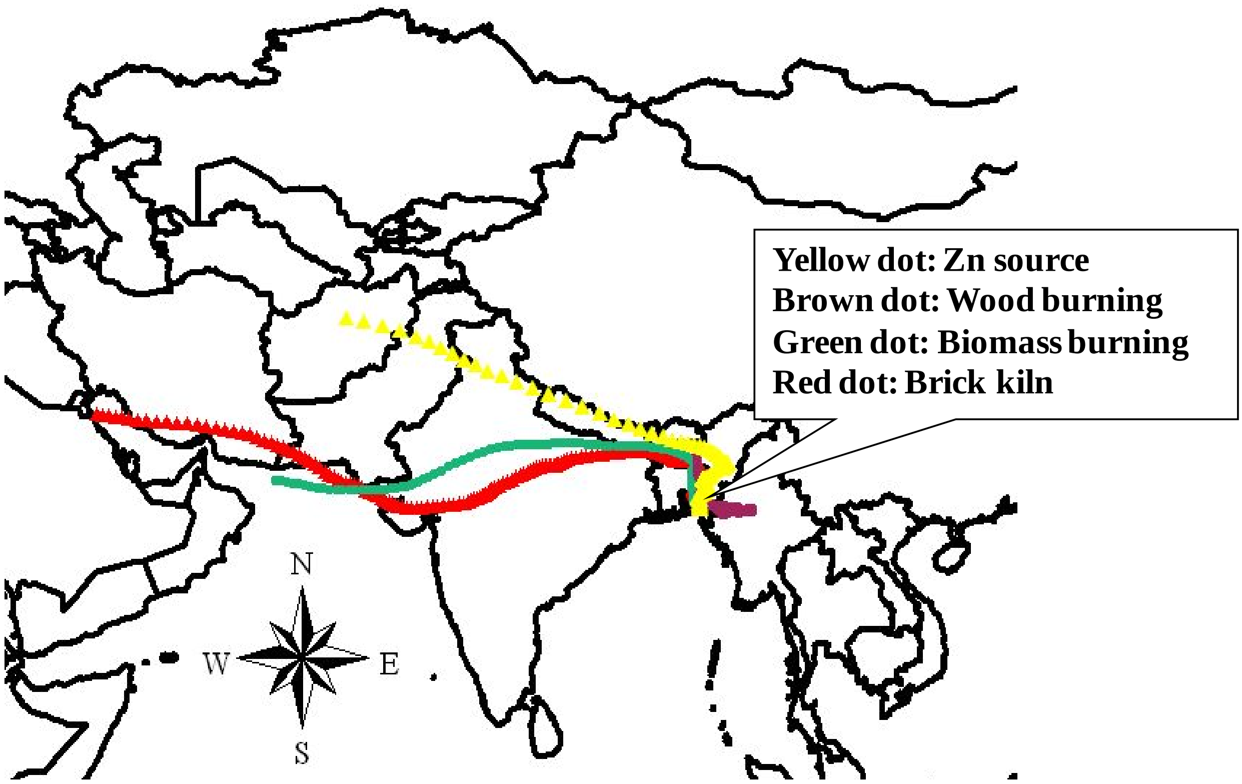

Figure 32: Back trajectory analyses for different source sectors indicating where air masses were transported before arriving at Chittagong

4 Impact of policy adaptation

4.1 Rajshahi city:

The results of the source apportionment results of Rajshahi city are given in Table 4 and compared with the previous PMF analysis results at the same location for a smaller data set covering the periods of 2001-2002 (Begum et al., 2004) for Rajshahi city. Previous source apportionment results (Begum et al., 2004) showed that brick kilns normally produced about 50% of fine particles ($PM_{2.5}$ particles).

The recent study shows that brick kilns and wood burning produce about 75.6% of fine particles. The concentrations of BC from these sources is also high. The current source apportionment data (Table 4) shows that the contribution of BC from motor vehicles has decreased since the previous study (2004) following adoption of CNG vehicles in 2003. Air quality policy actions were taken leading to PM and BC emissions reductions from motor vehicles.

Table 4: Sources factors for fine PM from Rajshahi aerosol study in different years

Source	2001-2002			2010-2012			
	%	Mass	BC/EC	%	Mass	BC/EC	Delta-C
		µg/m3			µg/m3		
Soil dust	1.88	0.37	0.00	8.39	10.7	0.00	0.00
Road dust	5.29	1.06	0.00	2.91	3.69	0.07	0.06
Sea salt	13.89	2.77	0.00				
CNG							
Brick kiln	50.4	10.08	4.62	40.2	51.0	6.32	3.34
Wood burning				35.4	45.0	1.95	0.27
Motor vehicle	28.5	5.69	1.18	9.80	12.4	3.44	0.31
Fugitive Pb				3.28	4.17	0.27	0.13
Reconstructed mass		**20.0**	**5.80**		**127**	**12.1**	**4.13**
Measured mass		**22.9**	**6.34**		**149**	**13.0**	**5.68**

4.2 Dhaka city:

The most recent source apportionment results for Dhaka city are given in Table 5. It has been found that the contribution from motor vehicles as well as BC concentrations have decreased since the previous study in 2004. This result is a positive achievement for the policies adopted by the Government. GDP growth in Dhaka has been stagnant, but the growth in the number of motor vehicles has continued [27]. However, the contributions of fine PM and as well as BC concentrations have decreased.

CNG powered vehicles are playing a positive role in economy of the country. Average CNG usage is 92.19 MMCM per month, which is equivalent to 0.065 million litres of petrol/octane. Bangladesh imports about 1.2 million metric tons of crude oil along with 2.6 million metric tons on refined petroleum products per annum. The major consumer of liquid fuel is transport, followed by agriculture, industry, and commercial purposes. Since the price of CNG is much lower than other fuels, it has been widely adopted. The Government has also decided to ban motorized rickshaws in many parts of Dhaka, without improving public transport, walking, and bicycle riding facilities. As a result, the demand for private cars has increased with vehicular number growth of more than 10% per year (BRTA, 2012). There have also been changes in the type of the vehicles including the reduction in new two-stroke vehicles, conversion of buses to compressed natural gas, and retirement of old vehicles.

At present, there are five types of brick kiln technologies existing in Bangladesh including Bull Trench Kilns (BTK), Fixed Chimney Kilns (FCK), Hybrid Hoffman Kilns (HHK), Zigzag Brick Kilns (ZBK) and Vertical Shaft Brick Kilns (VSBK). Table 6 shows different types and numbers of brick kilns in Bangladesh. Because of limited availability of gas resources, the Hybrid Hoffman Kilns that are using natural gas as a fuel are limited. The BTKs are not permitted by the government because of their high emissions. They were replaced by FCK in the past. Because of relatively low investment cost, FCK became the most popular brick kiln technology in Bangladesh with 92% of share of total existing brick kilns. However, it also has serious emissions problems [28]. The high fixed chimney does not reduce the carbon emissions due to use of very low level firing technology. As a result, FCKs emit a huge amount of particulate matter (PM) as well as other flue gases. The government has decided to impose a ban on FCK to reduce their environmental impacts. Recognizing the importance of reduction air pollution through improvement of energy efficiency as well as reducing emissions from brick kilns; the Bangladesh Government has been trying to improve the brick kiln technologies in various ways. New brick kiln technologies have recently been introduced including Hybrid Hoffman Kiln using coal as fuel in place of gas, continuous Vertical Shaft Brick Kiln (VSBK), Tunnel Kiln, etc. Those kilns are now being operated on an experimental basis at a limited scale.

However, because of the increased number of brick production industries, the emissions from brick kilns has become larger than any other PM source [29]. The contribution of BC from the brick kilns is even higher than the contributions from motor vehicles (Begum et al., 2013). The recent data set (2010-2012) shows that BC emission has been reduced relative to the previous year. Thus, the relatively limited rise in fine PM concentrations (considering measured mass in different year) indicates that control actions have helped to balance the increases in pollution that would have been anticipated to parallel the growth in population, economic activity, and the number of vehicles.

A separate issue of concern for Dhaka is the increase in Pb concentrations. The relative contribution of Pb has more than doubled since the previous study (from 3.77% in 2001-2002 to 7.63% in 2010-2012.

Table 5: Source factors for fine PM in the Dhaka aerosol study in different years

Source	**2001-02**			**2010-11**				**2010-12**			
	%	**Mass**	**BC/EC**	**%**	**Mass**	**OC**	**BC/EC**	**%**	**Mass**	**BC/EC**	**Delta-C**
		ug/m3			**ug/m3**				**ug/m3**		
Soil dust	1.00	0.59	0.00	5.28	4.03	0.78	0.14	7.57	3.98	ear0.95	0.00
Road dust				8.62	6.59	1.91	1.92	7.70	4.05	0.63	0.00
Sea salt				5.78	4.42	0.26	0.05	1.33	0.70	0.01	0.00
Diesel				**22.1**	**16.84**	**4.73**	**1.54**				
Gasoline				**7.06**	**5.39**	**1.40**	0.75				
Brick kiln	**37.5**	**22.4**	**13.0**	**40.8**	**31.18**	**7.91**	**5.99**	**58.0**	**30.5**	**3.20**	**1.52**
Biomass burning								7.37	3.87	1.11	0.72
Zn source	2.41	1.44	0.45								
Motor vehicle	**43.0**	**25.7**	**11.8**					10.4	5.49	0.03	0.07
Unknown source	12.7	7.60	0.0								
Fugitive Pb	3.32	1.98	0.00	10.4	7.93	3.77	1.98	7.63	4.01	0.18	0.00
Reconstructed Mass		59.7	25.2		76.4	20.8	12.4		52.5	6.12	2.32
Measured mass		71.7	27.7		**85.1**	**23.3**	12.8		63.9	7.11	2.96

Table 6: Existing brick kiln technologies in Bangladesh

Type of Kiln	Number	Total kiln (%)	Brick production (in billions)	Total production (%)
Fixed Chimney Kiln	≤4500	92	15.8	91.4
Zigzag Brick Kilns	≤150	3	0.6	0.0
Hoffmann (Gas)	≤20	0.4	0.2	3.5
Hybrid Hoffman Kilns	≤10	0.2	0.2	1.4
Others	≤200	4	0.5	0.9
Total	4880	100	17.5	100

4.3 Chittagong city:

From Table 7 (Chittagong), it can be seen that the fine PM mass contribution from brick kilns has increased slightly from the 2006-07 period. However, the BC contributions have decreased [30]. The second largest contribution comes from a Zn source factor.

In Chittagong, there are many industries where Zn (Pb is added to improve the reflectance) is used for electroplating. The other Pb source is the battery industries and also from the secondary Pb smelter where rejected batteries are recycled to make new Pb batteries; Pb levels for Chittagong in the current study are similar to the current levels in Dhaka.

CNG as a source factors contributed considerably in 2006-07.This source factor was not identified in 2010-12. It is believed that because of increased CNG adoption the contribution from motor vehicles generally has decreased.

Table 7: Source factors of fine PM from the Chittagong aerosol study in different years

Source factor	2006-07			2010-12			
	%	Mass	BC/EC	%	Mass	BC/EC	Delta-C
		µg/m3			µg/m3		
Soil dust	13.6	5.37	0.96	2.59	1.65	0.02	0.00
Road dust	2.05	0.81	0.78	1.54	0.98	0.01	0.00
Sea salt	1.00	0.39	0.34				
CNG	16.2	6.39	0.00				
Brick kiln	***35.5***	***14.0***	***5.36***	***36.2***	***23.1***	***3.03***	***1.21***
Wood burning				19.1	12.2	0.36	0.60
Zn source	21.8	8.60	0.00	30.5	19.4	1.92	1.00
Motor vehicle	***9.76***	***3.85***	***2.65***	***2.53***	***1.61***	***1.25***	***0.03***
Fugitive Pb				7.44	4.73	0.49	0.14
Reconstructed mass		**39.4**	**10.1**		**63.7**	**7.08**	**2.98**
Measured mass		**45.9**	**11.3**		**73.3**	**7.59**	**3.41**

4.4 Khulna city:

Table 8 represents the apportionment of sources from fine particles during the sampling period for all sites. It is observed that the contribution of brick kilns including wood burning or biomass burning is higher in Rajshahi than at the other three sites. The same has been observed in case of motor vehicle sources.

The anthropogenic activities in Khulna city in general are less than in other cities but the fine PM concentrations as well BC are similar to levels seen in Dhaka where those activities are much higher; this could be due to local sources close to the site in Rajshahi influencing the results. However, during winter months, the wind blows from north-west towards south-west direction, which as a result, the regional transported air pollutants could also be increasing the particle concentrations observed in Khulna city.

Table 8: Source factors of fine PM based on the sampling undertaken during the study period 2010-2012

Source	Rajshahi				Dhaka				Khulna				Chittagong			
	Contri-bution	Mass	BC	Delta-C	Contri-bution	Mass	BC	Delta-C	Contri-bution	Mass	BC	Delta-C	Contri-bution	Mass	BC	Delta-C
	(%)	ug/m3			(%)	ug/m3			(%)	ug/m3			(%)	ug/m3		
Brick kiln	40.20	51.0	6.32	3.34	58.0	30.5	3.20	1.52	36.1	16.1	1.78	1.78	36.2	23.1	3.03	1.21
Biomass burning					7.37	3.87	1.11	0.72	3.60	1.61	0.17	0.17				
Road dust	2.91	3.69	0.07	0.06	7.70	4.05	0.63	0.00	9.75	4.36	1.50	1.50	1.54	0.98	0.01	0.00
Soil dust	8.39	10.7	0.00	0.00	7.57	3.98	0.95	0.00	9.00	4.03	0.00	0.00	2.59	1.65	0.02	0.00
Motor vehicle	9.80	12.4	3.44	0.31	10.4	5.49	0.03	0.07	13.7	6.12	0.22	0.22	2.53	1.61	1.25	0.03
Fugitive Pb	3.28	4.17	0.27	0.13	7.63	4.01	0.18	0.00	8.05	3.60	0.39	0.39	7.44	4.73	0.49	0.14
Sea salt/ Zn source					1.33	0.70	0.01	0.00					30.5	19.4	1.92	1.00
Wood burning	35.4	45.0	1.95	0.27					19.9	8.91	1.10	1.10	19.1	12.2	0.36	0.60
Reconstructed mass		127	12.1	4.13		52.5	6.12	2.32		44.8	5.17	1.03		63.7	7.08	2.98
Measured mass		149	13.0	5.68		63.9	7.11	2.96		59.4	5.60	1.57b		73.3	7.59	3.41

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ABSTRACT
This study is the first preliminary analysis of PM and BC data under Task 3 of the Bangladesh Air Pollution Studies (BAPS) project. The basis of Task 3 work is outlined in the report Task 3 (Source Apportionment) Inception Report prepared by NILU June 2012.
Discussions of results from four cities in Bangladesh is included in the report. More than 62% of the PM2.5-10 was soil and road dust in Rajshahi, Chittagong and Khulna sites but in Dhaka, the dust contribution was about 38%.
From the Dhaka data, PMF modeling resolved 7 sources for the coarse fraction PM samples. The identified sources are brick kilns, road dust, motor vehicles, soil dust, sea salt, Zn source and fugitive Pb. Because of the brick production technology, bricks are produced during dry periods mainly starting from November to early March every year. This profile has seasonal variation and has high contribution in winter and also influenced by north-westerly wind.

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