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

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Authors' contributions

This work was carried out in collaboration between all authors. Author MN organized the sampling campaign and arranged the logistics for sample collection. Authors SR, BS, and PKH organized the analysis of samples. Author BAB performed the statistical analysis, and wrote the first draft of the manuscript and managed literature searches. Author MN is the Project Director of the CASE Project and assisted in coordinating the sampling process. Author PKH along with authors SR and BS assisted in the evaluation of the PMF results and their interpretation with respect to source identification and quantitative apportionment. All authors read and approved the final manuscript.

Original Research Article

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ABSTRACT

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 Sapura in Rajshahi, Farm Gate in Dhaka, Baira in Khulna and a TV station, Khulshi, in Chittagong. PM sampling was performed using dichotomous samplers, which collect samples in two sizes: $PM_{2.5}$ 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 PMF2 modeling. 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

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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. Hence in order to reduce the local particulate pollution, it will be necessary to take policy actions regionally.

Keywords: $PM_{2.5-10}$; $PM_{2.5}$; BC; Delta-C; long range transport.

1. INTRODUCTION

Currently, urban air pollution and its effects are an issue of great concern for developing countries. To address the air pollution issues, it important to know the possible sources and their strengths so actions can be taken that can effectively and efficiently improves air quality. Local sources can be controlled by local initiatives, but regional as well as transboundary issues would require intergovernmental interventions. In Bangladesh, particulate matter pollution is more severe than for gaseous pollutants. Several studies have shown that $PM_{2.5}$ (Particle size <2.5 micrometer) has significant negative impact on human health [1-3]. Previously source apportionment studies were performed for the same site of Dhaka city air pollution using data from June 2001 to June 2002 [4-6]. These source apportionment studies found that vehicles were normally responsible for about 50% of fine particles ($PM_{2.5}$ particles) in Dhaka. Coarse particles ($PM_{2.5-10}$ particles) (Particle size 2.5-10 micrometer) 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 [8]. Source apportionment based on the PM data can be used to examine the effect on these policy implementations. Several studies [6-9] have shown that there is also transboundary contribution during the wintertime when wind blows from north and northwest directions during winter.

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 PMF2 modeling for source apportionment in order to examine if there are significant changes in the source characteristics arising from the policy interventions.

2. MATERIALS AND METHODS

2.1 Sampling

Samples were collected on 37mm diameter Teflon filters using Thermo Andersen dichotomous samplers, which were programmed to sample at 16.7lpm for proper size fractionation. The samplers at each station were positioned with the intake upward and located in an unobstructed area at least 30cm from any obstacle to air flow. The sampler inlets were placed at a height of 10 m above ground level. Appropriate quality control/quality assurance (QA/QC) protocol was followed 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.

2.1.1 Site description and measurement period

Rajshahi, a metropolitan city, is situated in the northern region of Bangladesh (latitude 24.37ºN, longitude 88.70ºE) 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.

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

Khulna, the third largest city of the country, is situated in the southern region of Bangladesh (latitude 22.48N, longitude 89.53ºE) and near the Bay of Bengal. Being located in a large river delta, it is the second port area of Bangladesh. The CAM 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.

Chittagong (latitude 22.22ºN, longitude 91.47ºE) 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 2.5km 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 air pollutant concentrations of the city [11]. The major sources were biomass burning/brick kilns, soil dust, road dust, Zn source (including two-stroke motorcycles), motor vehicles, CNG (Compressed natural gas) vehicle, and sea salt in the Chittagong aerosol [11]. The PM samples were collected between September 2010 and July 2012 depending on the cities and the sampling time was from 10 am to 10 am (24h) (Table 1).

2.2 PM Mass and BC Analysis

PM mass was measured in the laboratory of the Department of Environment. 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 (22ºC) 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].

2.3 Multielemental Analysis

Multielemental analyses of the collected samples were made using X-Ray Fluorescence (XRF) using a Spectro X-LAB2000 spectrometer. Twenty six species determined in all including black carbon (BC) and delta-carbon (Delta-C) for each 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 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 has found that due to the missing of organic carbon, the least square fitting in case of the fine particle samples was not good as for the coarse particle samples.

2.4 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.

2.5 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.

2.6 Positive Matrix Factorization Modeling

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

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

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 are also presented from PMF modelling are presented in above Table 3. The detailed description of factor profile is described in Result and discussion section. 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.

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

Parameter	Rajshahi (24.38°N, 88.61°E)				Dhaka (23.76°N, 90.39°E)				Khulna (22.48°N, 88.61°E)				Chittagong (22.36°N, 91.80°E)			
	$PM_{2.5-10}$	$PM_{2.5}$	BC	Delta-C	$PM_{2.5-10}$	$PM_{2.5}$	BC	Delta-C	$PM_{2.5-10}$	$PM_{2.5}$	BC	Delta-C	$PM_{2.5-10}$	$PM_{2.5}$	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			

Table 2. The summary of least squares fit from RCM calculation to the measured mass in each site

Site	Coarse		Fine	
	Slope	Correlation coefficient	Slope	Correlation 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

Table 3. The summary of least squares fit from PMF modeling to the measured mass in 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

2.7 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 to 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 Figs. 1 and 2, respectively. Fig. 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 [4]. The contribution of this source is from both south-east and north-west direction. The coal that is burnt in kiln contains 4 to 6% sulfur. Due to 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 [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 [26] 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 [4] and has seasonal variation. This factor is highly influenced by southerly wind.

The fourth source profile has the characteristics of high Na, Mg, Al, Si, P, Cl, K, Ca, Ti, Mn and Fe and represents soil dust [5] and also influenced by west 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 [5] and has seasonal variation that is influenced by westerly winds. The diesel fuel in Bangladesh contains about 3000ppm sulfur. Heavy duty vehicles mostly use this fuel. However, heavy-duty diesel trucks can only be used in Rajshahi 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 [10] and shows no seasonal variation with few high peaks. The coarse Pb comes from battery reclamation.

Breaking up the batteries produce large particles where resmelting the Pb produces small particle Pb. The factor shows high contribution from south. 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. This factor is influenced by north-westerly wind. It has found the coarse fraction carries about 62.2% of soil dust including road dust.

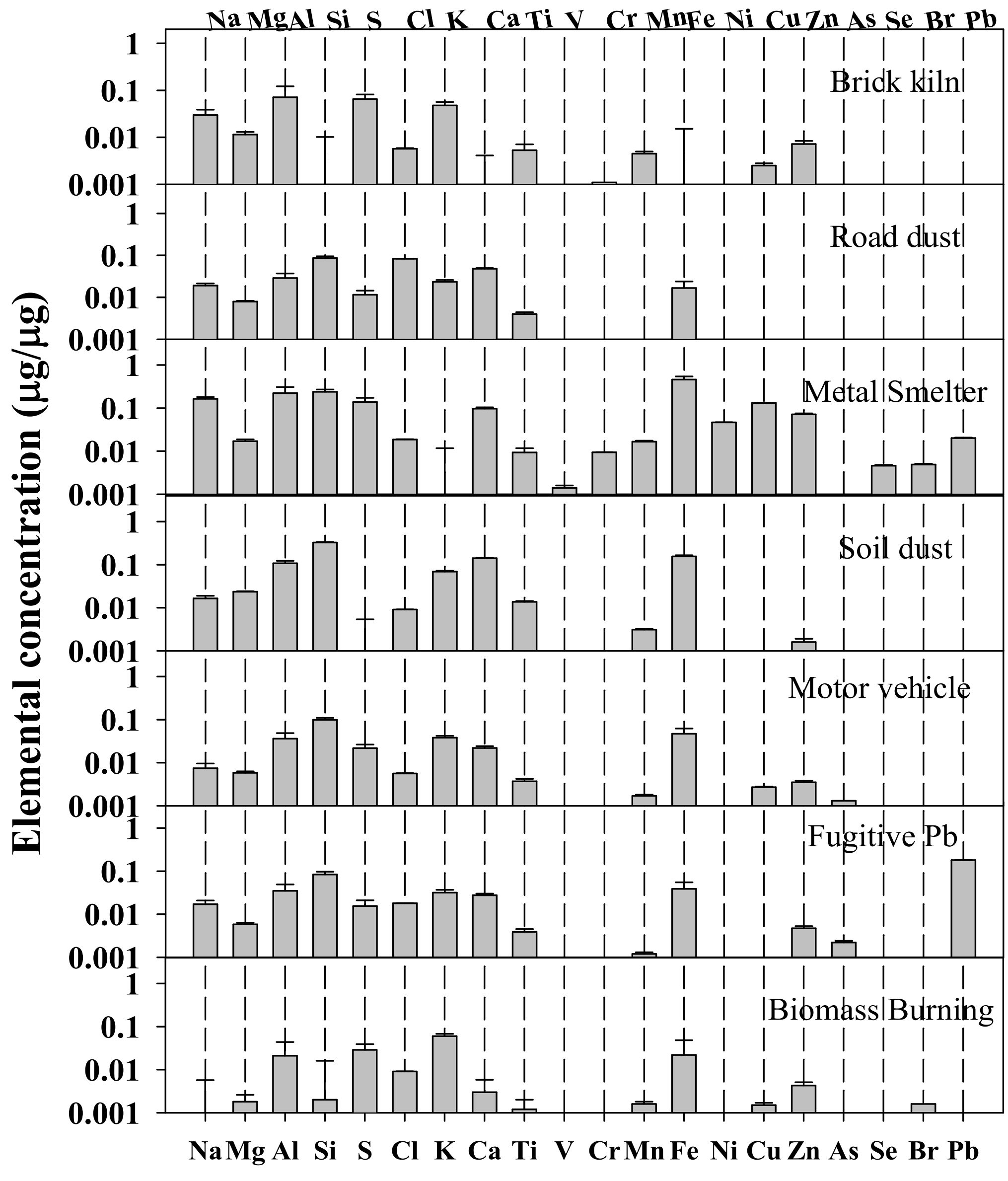


Fig. 1. The identified source profiles of each source for coarse fraction

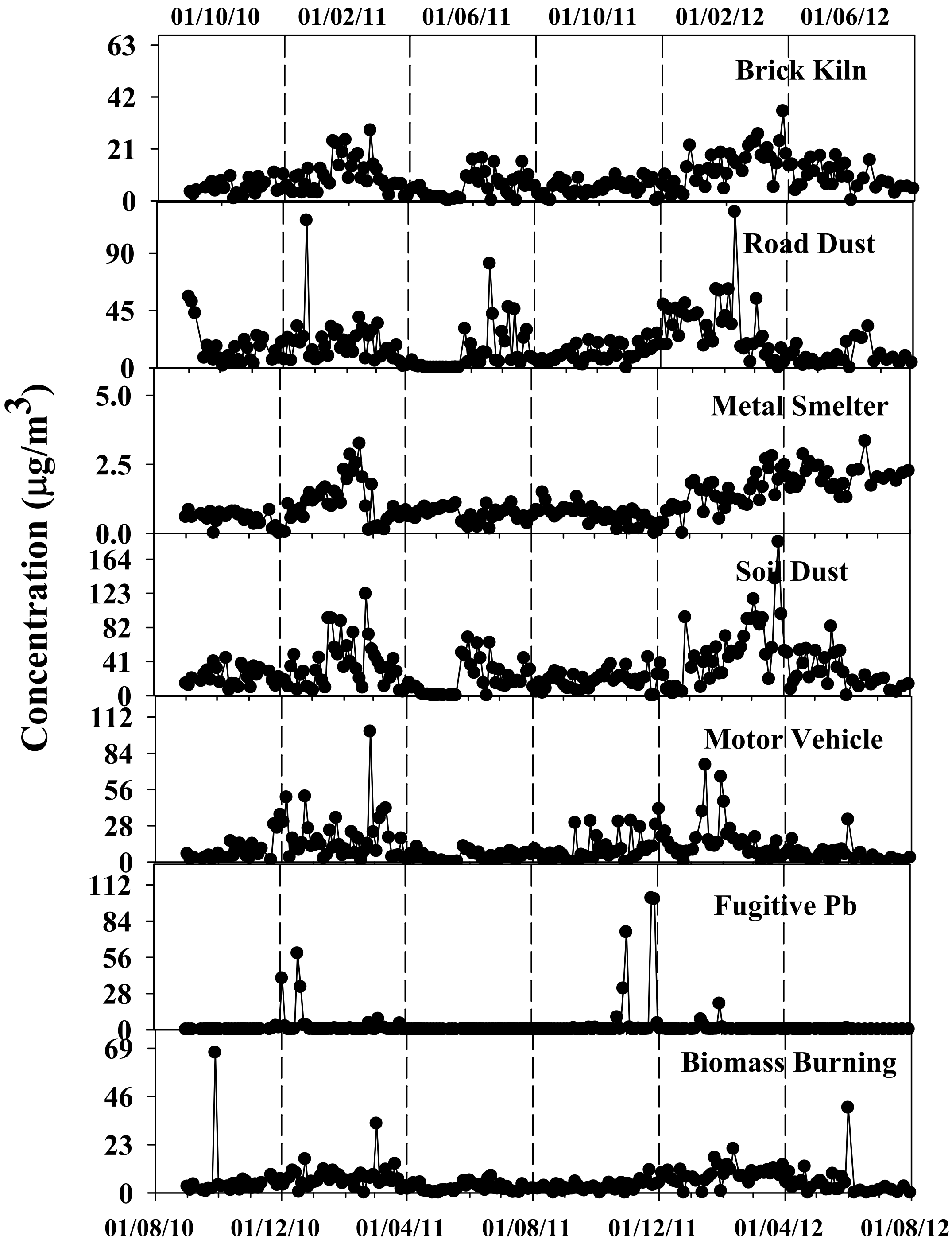


Fig. 2. The mass contribution of identified source of coarse fraction

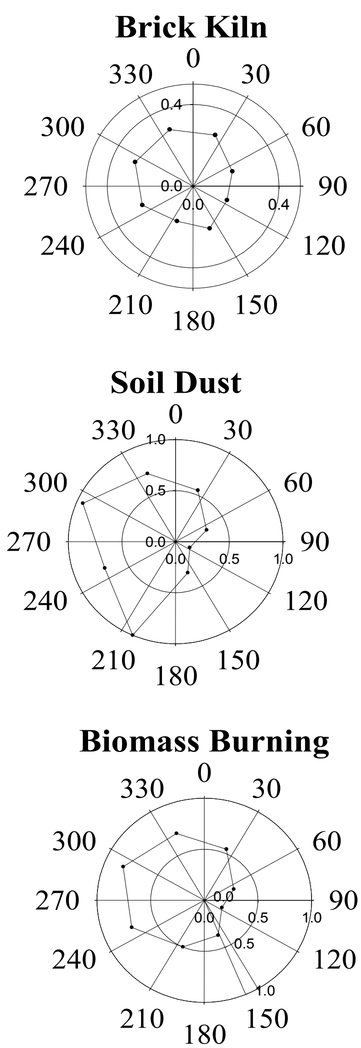


Fig. 3. The directional pattern of each source for coarse particle

From the fine PM data from Rajshahi, PMF modeling resolved six sources (Figs. 4 and 5) and the characteristics of sources are same as in coarse PM. The seasonal influence and the directional pattern (Fig. 6) are same as in coarse particles. The fine fraction carries about 85.4% of anthropogenic sources such as brick kiln, motor vehicle and wood burning sources. The regression slope and coefficient is for both coarse and fine fractions are given in Table 3. It has found that directional patterns of fine sources are same as in coarse. The seasonal wind directional pattern for Rajshahi city is shown in Fig. 7. From Fig. 5, it has found that the contribution of high fugitive Pb source, biomass burning and soil dust contributions are on 3 December 2010, 13 February 2011 and 18 March 2012, respectively. From Fig. 8, it is observed that due to the contribution of long range transport on those days, the overall pollution concentration has increased.

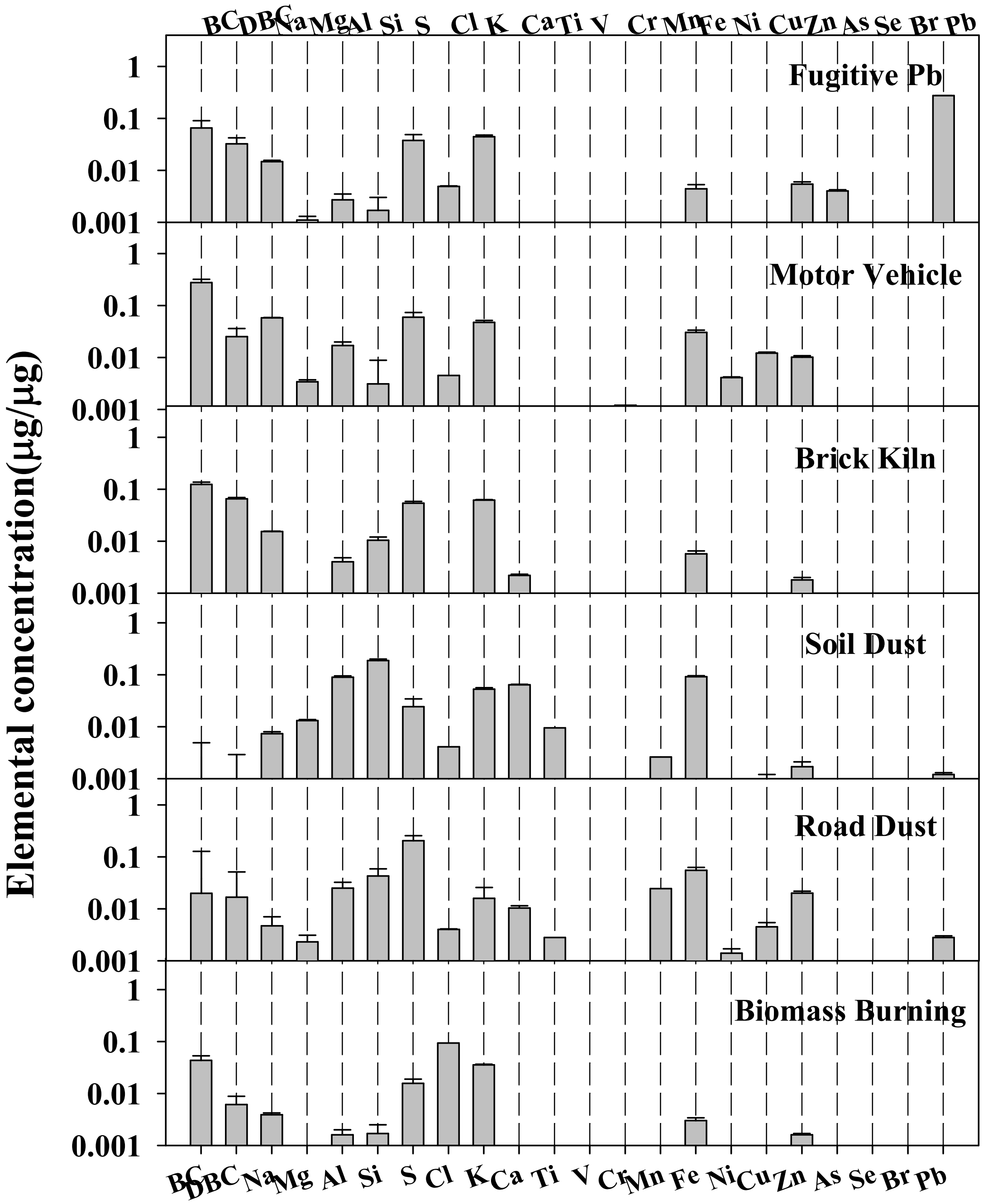


Fig. 4. The identified source profiles of each source for fine fraction

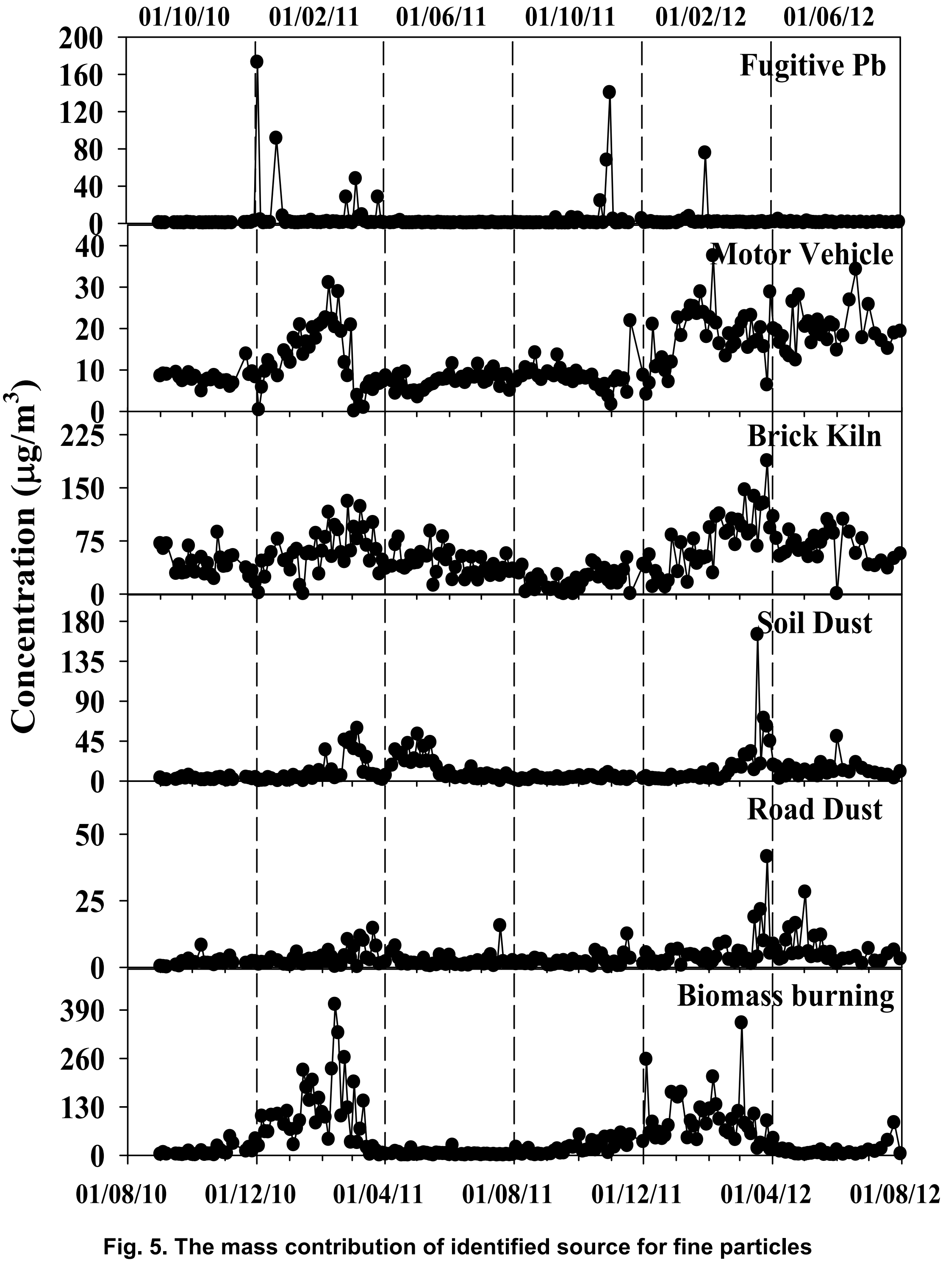


Fig. 5. The mass contribution of identified source for fine particles

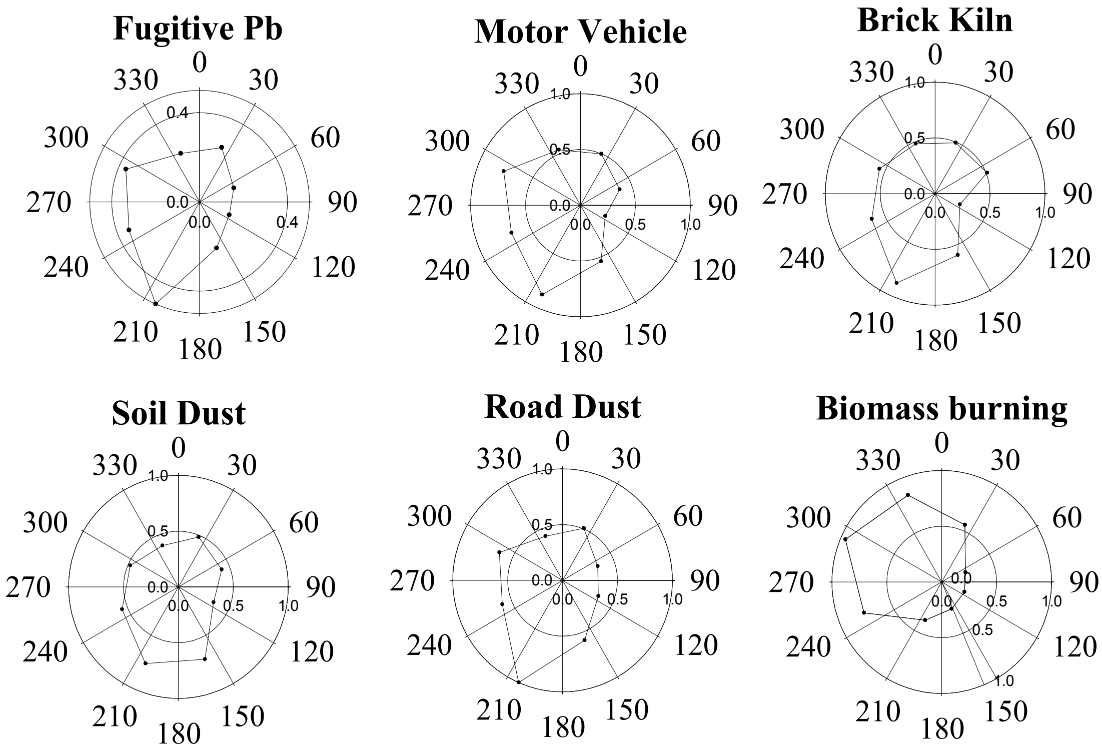
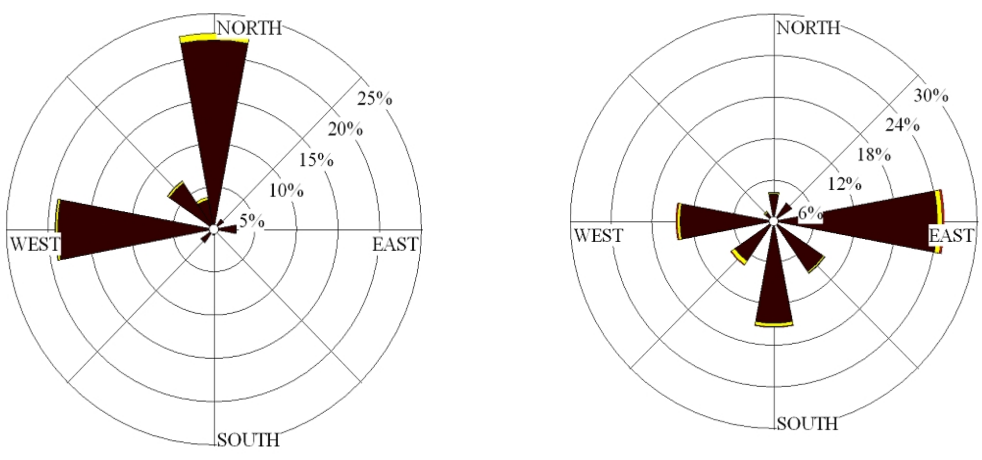


Fig. 6. The directional pattern of each source for fine particle



Winter

Pre-monsoon

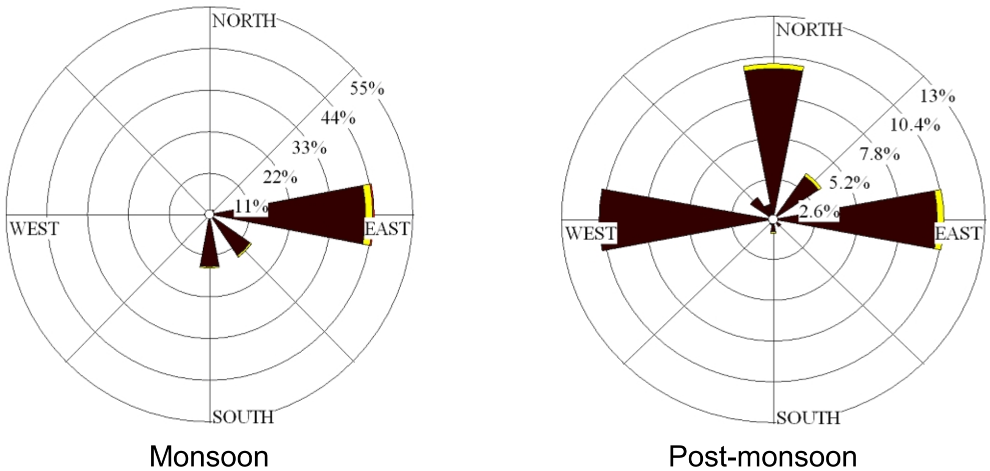


Fig. 7. Wind direction pattern at Rajshahi city

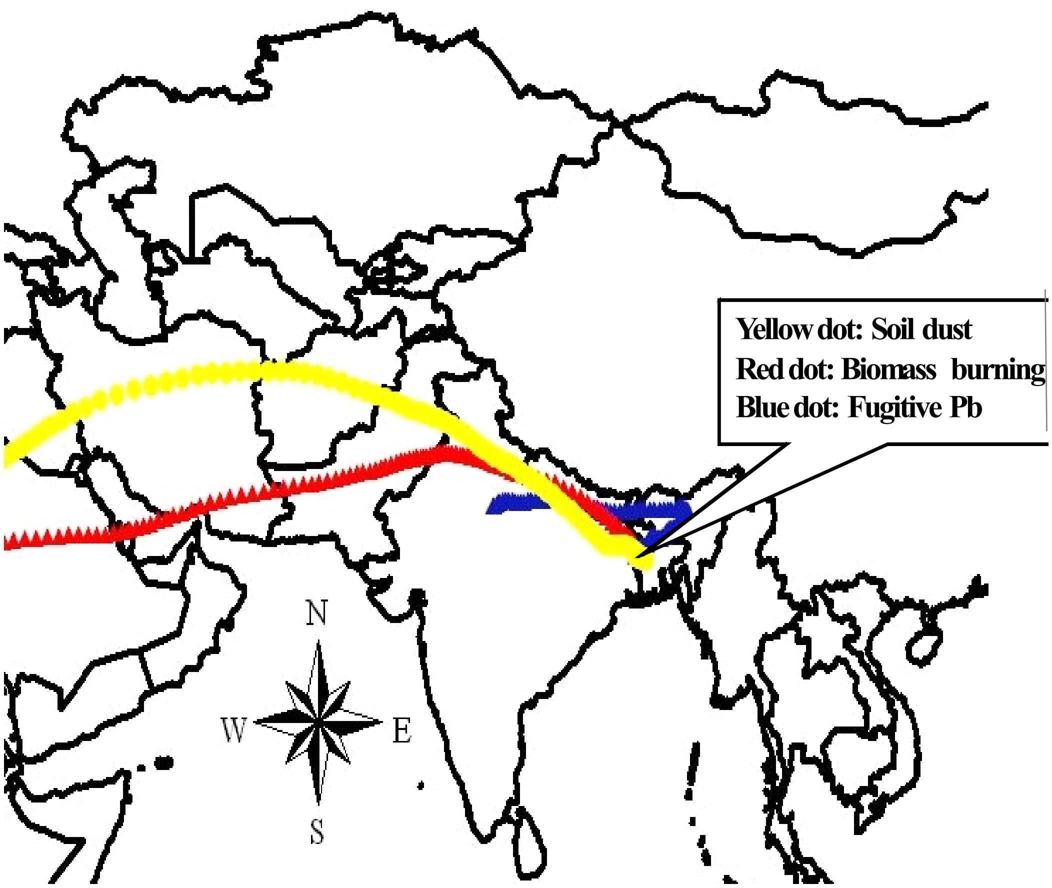


Fig. 8. Evidence of long range transport at Rajshahi city

3.1.2 Dhaka CAMS2 site

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. The sources have directional influence [17]. The first source has the characteristics of high Al, Si, S, Cl, K, Ca, and Fe and represents brick kiln. The coal that is burnt is 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 profile has seasonal variation and has high contribution in winter [6] and also influenced by north-westerly wind.

The second source profile has the characteristics of high BC, Al, Si, S, K, Ti, Mn, Cu, As, 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 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 Zn source. Zn may come from the galvanizing factories and to increase the reflectance properties, Pb is added during manufacturing [27]. 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 fine Pb is from battery recycling.

From fine PM data, PMF modeling resolved seven sources and the characteristics of sources are same as in coarse PM. The seasonal influence and the directional pattern [10] are same as in coarse particles. The fine fraction carries about 68.4% of anthropogenic sources such as brick kiln, motor vehicle and wood burning sources. The regression slope and coefficient is for both coarse and fine fractions are given in Table 3. It has found that directional patterns of fine sources are same as in coarse fraction results. The seasonal wind directional pattern for Dhaka city was discussed elsewhere [28]. From seasonal influence (Fine fraction) of Fig. 9, it can be seen that the contribution of high fugitive Pb source, biomass burning, and soil dust sources peak on 21 November 2010, 3 March 2011 and 14 April 2011, respectively. From Fig. 10, it has found that the contributions of long range transport on those days have increased the measured local pollution concentrations.

3.1.3 Khulna CAMS site

From the data of Khulna CAMS site, PMF modeling resolved 7 sources for both coarse and fine fractions of PM samples. The identified sources 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 characteristics of sources are similar to those discussed for the above two sites. The regression slopes and coefficients are of both coarse and fine fractions are given in Table 3. Fig. 11 shows wind directional pattern of Khulna city. Fig. 12 shows the long range transport of fine sources at Khulna site.

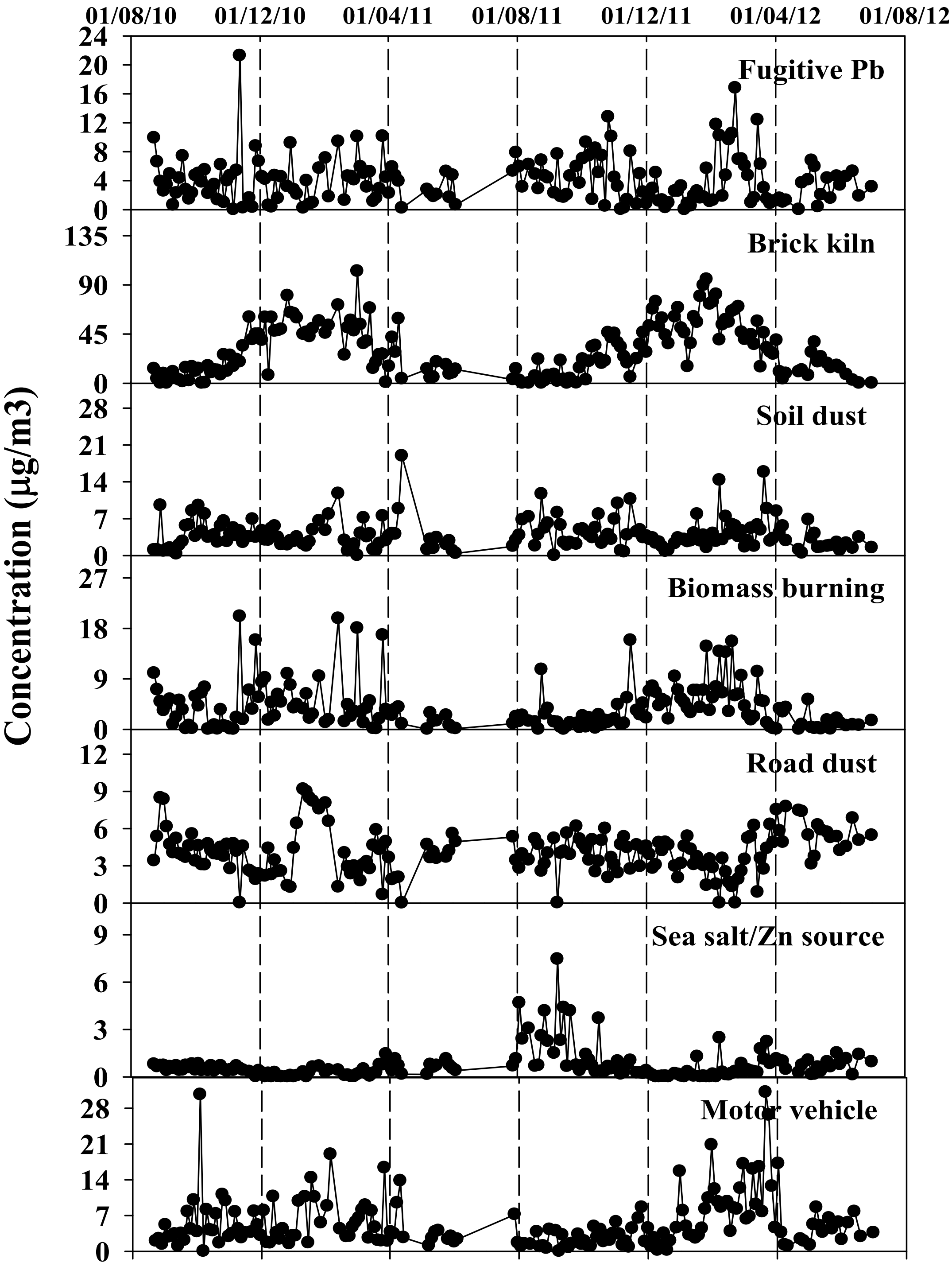


Fig. 9. The identified source profiles of each source for fine fraction at Dhaka

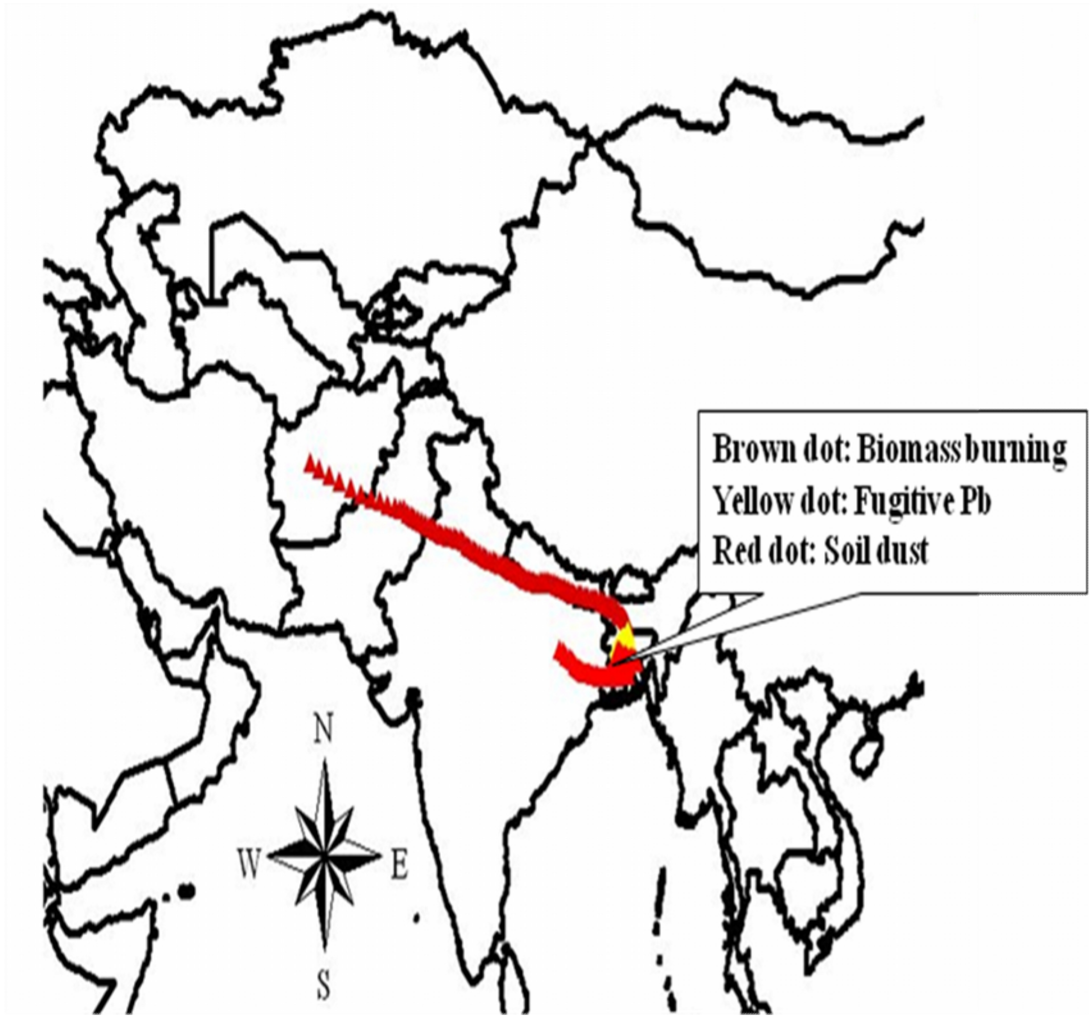


Fig. 10. Long range transport arriving at Dhaka

3.1.4 Chittagong CAMS site

From the data of Chittagong CAMS site, PMF modeling resolved 7 sources 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 fraction of PM. The characteristics of sources are similar to those discussed for other sites. The regression slope and coefficient is for both coarse and fine fractions are given in Table 3. Fig. 13 shows the long range transport of fine sources at Chittagong site.

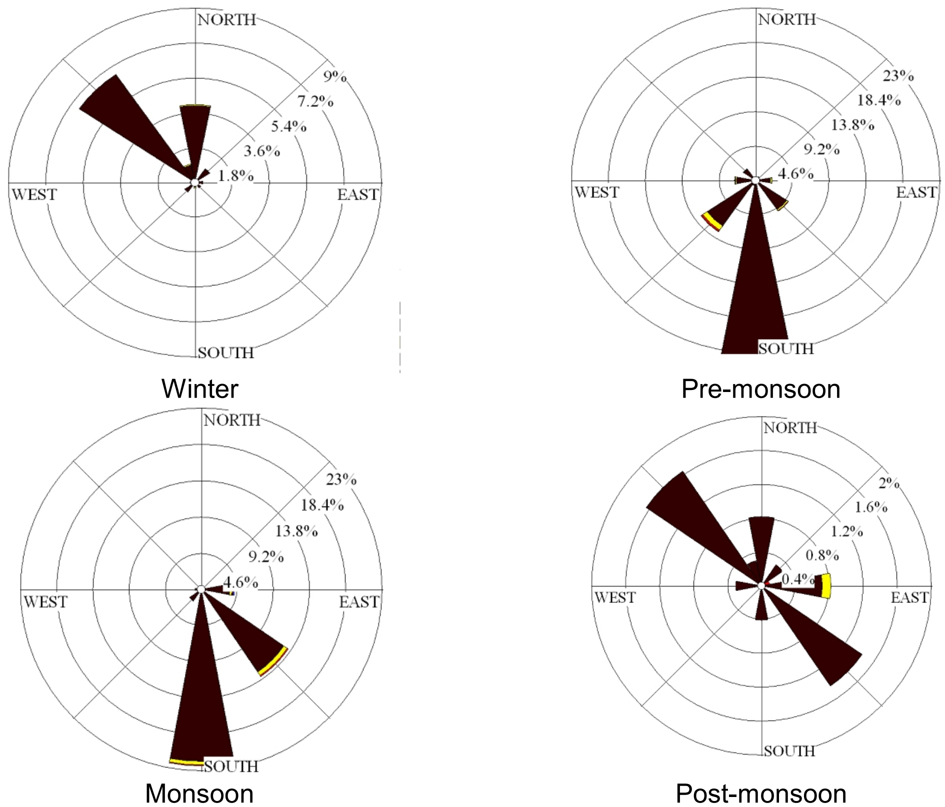


Fig. 11. Wind direction pattern at Khulna city

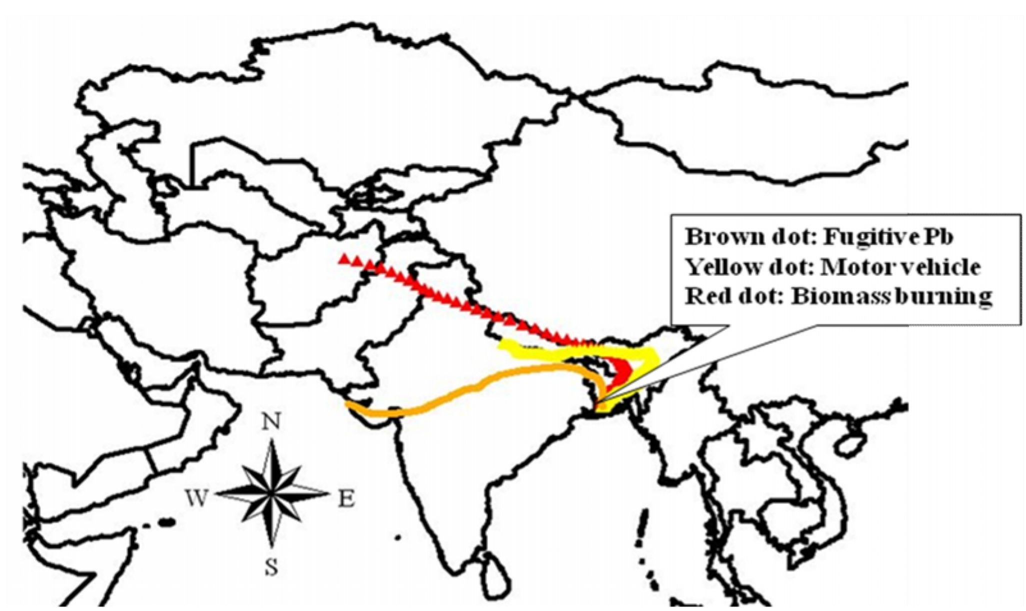


Fig. 12. Long range transport arriving at Khulna

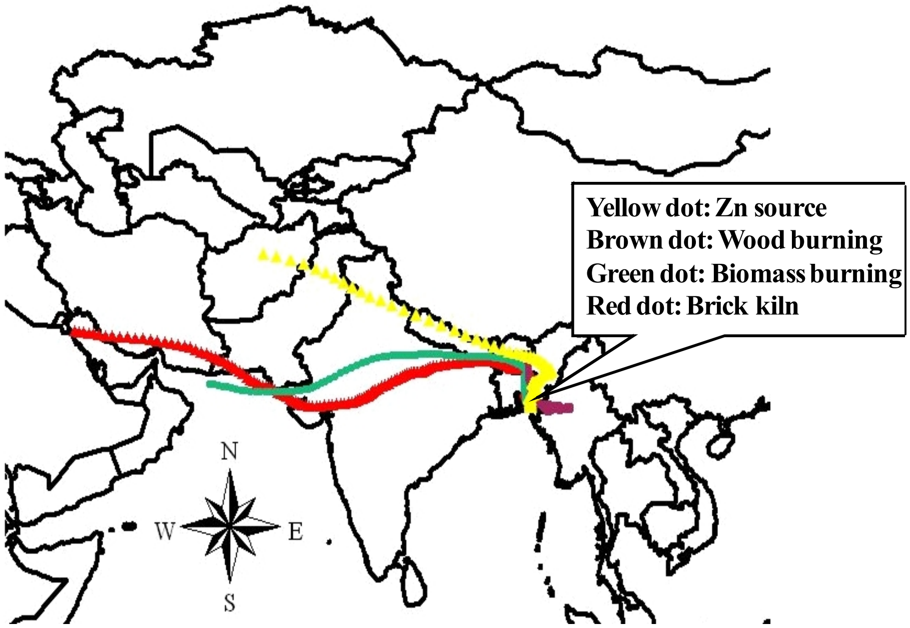


Fig. 13. Long range transport arriving at Chittagong

3.2 Impact of Policy Adaptation within Cities

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 [4] for Rajshahi city. The earliest source apportionment results [4] showed that brick kilns normally produced about 50% of fine particles ($PM_{2.5}$ particles). The recent study shows that brick kiln and wood burning produce about 75.6% of fine particles and emission of BC from these sources is high. The current source apportionment data (Table 4) shows that the contribution of BC from motor vehicles has decreased following adoption of CNG vehicles in 2003. Air quality policy actions were taken to produce PM and BC emissions reductions from motor vehicles.

The source apportionment results for Dhaka city are given in Table 5. It has found that the contribution from motor vehicle as well as BC has decreased than the previous year. 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 [29]. However, the contributions of fine PM as well as BC emissions 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 liters 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 nature 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. Except HHK, rest of the kilns use coal and wood as fuel. 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 [30]. 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 ban on existing 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 existing brick kiln technologies in various ways. New brick kiln technologies have recently been introduced including HHK using coal as fuel in place of gas, continuous 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 [31]. The contribution of BC from the brick kilns is even higher than the motor vehicles [17]. 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 vehicles.

Table 7 represents the apportionment of sources for Khulna city from fine particles during the sampling period. 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 observation has found in case of motor vehicle sources. The anthropogenic activities in Khulna city is less than other cities but the fine PM concentrations as well BC is same as Dhaka where those activities are much high in Dhaka. During winter months, the wind blows from north-west towards south-west direction. As a result, transported air pollutants increase the local pollutants of Khulna city. Hence, it may conclude that the high concentration at Khulna is due to sweeping of pollutants from up winds.

From Table 8 (Chittagong), it can be seen that the fine PM mass contribution from brick kilns has increased from the 2006-2007 period, but the BC contributions have decreased [32]. The second largest contribution comes from sea salt/Zn source. 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 acid batteries. Because of CNG adaptation, the contribution from motor vehicles has decreased.

Table 4. Sources (Fine PM) from Rajshahi aerosol in different year

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

Table 5. Sources (Fine PM) from Dhaka aerosol in different year

Source	2001-2002			2010-2011				2010-2012			
	%	Mass	BC/EC	%	Mass	OC	BC/EC	%	Mass	BC/EC	Delta-C
		μg/m3				μg/m3				μg/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

Table 7. Sources (Fine PM) from Chittagong aerosol in different year

Source	2006-2007			2010-2012			
	%	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

Table 8. Sources from Fine PM during study period

Source	Rajshahi				Dhaka				Khulna				Chittagong			
	Contribution (%)	Mass	BC	Delta-C	Contribution (%)	Mass	BC	Delta-C	Contribution (%)	Mass	BC	Delta-C	Contribution (%)	Mass	BC	Delta-C
		μg/m3				μg/m3				μg/m3				μg/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	1.57		73.3	7.59	3.41

4. CONCLUSION

Based on particle compositional data for two size fractions of the atmospheric aerosol ($PM_{2.5}$ and $PM_{2.5-10}$), sources of the particles were identified and their contributions to the PM concentrations observed in the four largest cities of Bangladesh. All of these cities have high PM concentrations that do not meet the country's ambient air quality standards. There are common sources across the country including motor vehicles, road dust, biomass combustion, and brick kilns. There are clearly several locations with uncontrolled emissions from battery reclamation activities with resulting ambient concentrations well in excess of the air quality standard. Further control of local sources will be necessary if the air quality standards are to be achieved.

There is strong evidence for significant contributions of PM arising from long-range transport during the winter period. Further study of the role of this transported aerosol will be needed to assess the source locations and contributions in more detail.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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