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Technical Information

Trend Characteristics of Atmospheric Particulate Matters in Major Urban Areas of Bangladesh

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ABSTRACT The urban areas of Bangladesh suffer from severe air quality problem especially in dry season (November–April) when the PM concentrations frequently rise to 7–8 times of the WHO guideline value. The Clean Air and Sustainable Environment (CASE) Project of the Department of Environment has deployed countrywide continuous air monitoring systems to regularly monitor the air quality of the urban areas of Bangladesh. In this paper hourly concentrations of PM_{10} and $PM_{2.5}$ captured using β-attenuation method from 2013 to April 2018 in six important cities located in different regions of the country were exhaustively analyzed. Statistical analyses, diurnal and seasonal trends, and polar plots of PM concentrations were examined. Long range sources were spotted by Concentration Weighted Trajectory (CWT) method, where the trajectories were calculated using HYSPLIT-4 model. The analyses identified cities in the middle of the country (Dhaka, Narayanganj, Gazipur) as the most polluted ones while the city to the northeast region (Sylhet) was the least polluted. Average PM_{10} concentrations at Dhaka, Chattogram, Narayanganj, Gazipur, Sylhet and Barisal stations in dry seasons (November–April) were found 238.7±155.4, 190.7±108.5, 303.6±161.4, 227.3±142.7, 151.7±105.0 and 170.7±108.4 μg m^{-3} respectively whereas those in wet seasons (May–October) were 75.0±51.6, 55.5±40.8, 102.4±84.4, 60.6±48.5, 52.7±38.3, and 54.4±41.6 μg m^{-3} respectively. Correlative study of diurnal variations in PM concentrations and meteorological parameters revealed that the congenial meteorology aided in developing higher concentrations of both PM_{10} and $PM_{2.5}$ during nighttime. Sources located to the northwestern districts (Naogao, Rangpur, Bogura) were traced by the CWT method contributing to the air pollution in other regions of the country. Outside the boundary, sources in Nepal, and Delhi-NCR and Uttar Pradesh regions of India could have contributed to fine particles at the middle of the country.

KEY WORDS Particulate matter, Bangladesh, Conditional bivariate probability function, Diurnal variation, Concentration weighted trajectory

1. INTRODUCTION

The south Asian countries are now the major hotspots in the world for increased level of particulate matter (PM) in air (Pant *et al.*, 2018; Khatum *et al.*, 2017; Rana *et al.*, 2016a; Dey *et al.*, 2012; Gurung and Bell, 2012). Burning of biomass and fossil fuel (Tiwari *et al.*, 2014), dusts and industrial emissions fill the atmosphere with great amount of PM (Sharma *et al.*, 2016; Begum *et al.*, 2010; Sharma *et al.*, 2010;

Chowdhury *et al.*, 2007), and especially during dry season congenial meteorology assists in developing PM concentrations in the atmosphere (Schnell *et al.*, 2018). The heightened level of PM in air is not only increasing health burden in this region but also decreasing crop production and playing important roles in climate change (Pommier *et al.*, 2018; Tiwari *et al.*, 2014; Ramanathan and Carmichael, 2008; Venkataraman, 2005). Specially, the potential of black carbon in changing climate is now proven (Kopacz *et al.*, 2011; Shindell *et al.*, 2011; Yasunari *et al.*, 2010; Ming *et al.*, 2008). The State of Global Air/2019 published jointly by the United States' Health Effect Institute and Global Burden of Disease project of the Institute for Health Matrix and Evaluation reveals that air pollution could be blamed for about 4.9 million early deaths worldwide in 2017 (State of Global Air, 2019); China and south Asian region shared about half of this mortality. Lelieveld *et al.* (2015) also accounted outdoor air pollution for the early deaths of about 92,000 people in Bangladesh in 2010, 55% of which was attributed to the household energy use like cooking, heating, etc.

The country of Bangladesh has been experiencing continuous growth in urbanization as a result of ongoing industrial expansion. Urban population estimated at the last national consensus in 2011 was found about 28% of the total population, about 3.5% high from that found in the previous consensus in 2001 (BBS, 2011). The economic activities of the country have accordingly been changing from agriculture sector (contribution to GDP falls by 1.9% in last four years) to industrial sector (contribution to the GDP increases by 3.29% in last four years). Source apportionment of the air pollution in the cities is also changing gradually (Begum *et al.*, 2013, 2011, 2008). Vehicle sector contributed much (~50%) of the fine particles in Dhaka city in the 90's; but its share fell sharply as a result of the government's decisions of importing unleaded petroleum from 1999, complete expulsion of 2-stroke baby taxis in 2003, and introducing cleaner fuel compressed natural gas (CNG) in the vehicle sector. The latest apportionment result shown by CASE (2014) found brick manufacturing sector now contributing more than 50% of the fine particles in Dhaka, the capital city of the country. Not only Dhaka other divisional cities also suffer greatly from the emissions from brick kilns (CASE, 2014). Other than brick kilns, wood burning was found as a big source of fine particulates in Rajshahi, Khulna and Chattogram cities, and sea salt/zinc sources with contribution of about

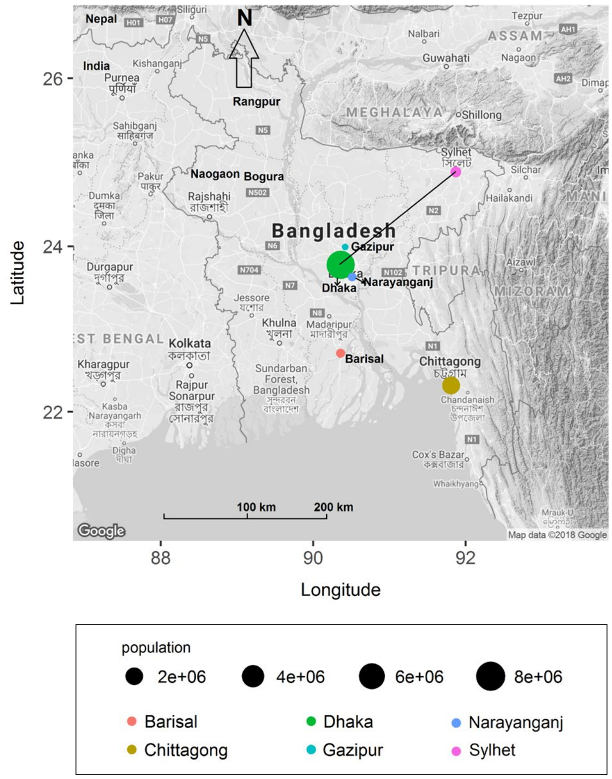

Fig. 1. Locations of air monitoring sites with population sizes of the respective town/city.

30.5% were found another strong source of fine particulates in Chattogram (CASE, 2014).

Although the entire country plunges into devastating air pollution during dry season, rare works have been done on monitoring and characterizing the air quality outside the Dhaka city. This study undertook massive initiatives of continuously capturing particulate matter (PM) concentrations in the air of 06 important cities located in different regions (center, south, southeast and northeast) of the country from 2013 to April 2018. The main objectives of this study were to characterize the temporal and spatial variations in atmospheric particulate matter concentrations in the country, and also to predict source directions responsible for high PM pollution. Seasonal, diurnal and directional trends and statistical parameters of PM_{10} and $PM_{2.5}$ concentrations were analyzed. Possible long range source-regions for the high PM concentrations in the cities were also investigated. This sort of countrywide observations and analyses of air quality were not done before in Bangladesh and hence carry a great deal of importance for the air quality management in the country.

Table 1. Descriptions of the cities, sampling sites, and possible sources of air pollution.

City, area and population, topography	Sampling site	Possible sources of air pollution in the city
Dhaka Capital and megacity; area ~316 km^2, population ~10.0 million, Flat	Coordinates 23.78N; 90.36E, sample intake ~9.0 m above the ground. Main city road ~100 meter away.	Thousands of brick kilns around the outskirts of the city, congested high emitting vehicle fleet, lots of small-scale factories, road dusts, open burning, etc.
Gazipur ~25 km to the north from Dhaka city; 47.23 km^2, 0.21 million, Flat	23.99N; 90.42E, sample intake ~9.0 m above the ground. 15 m away from a local road carrying low traffic. The site is relatively unaffected by nearby air pollution sources.	Lots of brick kilns are around, small-scale factories (Battery, garments, dye, etc.), road dusts, open burning, etc.
Narayanganj ~23 km to southeast from Dhaka; 47.23 km^2, 0.29 million, Flat	23.63N; 90.51E, sample intake at ~ 9.0 m above the ground. A road intersection ~1.0 km west from the site; diesel run water vessels run on the Shitalakkha river only 200 m south from the site	Industrial town having jute processing plants, paper and cement factories, and lots of small to medium industries like textile and dye factories, steel mills. Congested vehicle fleet and dusts and brick kilns.
Chattogram ~215 km to the southeast from Dhaka; 155 km^2, 2.6 million, hilly	22.32N; 91.81E, sample intake at ~ 9.0 m above the ground. Residential site not much influenced by local sources.	Port city with huge congested traffic, diesel vehicles and significant number of two-stroke baby taxis; brick kilns, heavy industries, steel mills, and natural sea salt.
Sylhet ~200 km to the northeast from Dhaka; 42.0 km^2, 0.53 million, hilly	24.89N; 91.87E, sample intake at ~14.0 m above the ground. The river Surma is 20 m away to the south. Two roads pass the site ~12 m to the east and south.	Vehicles and brick kilns.
Barisal ~120 km to the south from Dhaka; 69.0 km^2, 0.34 million, Flat	22.71N; 90.36E, sample intake at ~8.0 m above the ground. A road with low traffic passes the site.	Barisal is a big river port, large number of diesel vessel operates in the port; small number of vehicles, brick kilns.

2. MATERIALS AND METHODS

2. 1 Monitoring Sites

Continuous PM monitoring in air has been done in six important cities located at different regions of the country - Dhaka, Narayanganj and Gazipur in the middle, Barisal to the south, Chattogram to the southeast, and Sylhet to the northeast part of the country (Fig. 1). Table 1 provides information of the sampling sites and possible sources of air pollution in the respective cities.

2. 2 PM Monitoring, Quality Control and Analyses

Continuous measurement of PM concentrations at the sites was done using beta attenuation method by the PM monitors (Model BAM-1020) made by the Met One Instrument Inc, USA. In the monitor, Carbon-14 was used as the beta radiation source and Geiger Mueller (GM) counter as the detector for the beta particle counts. Individual monitors were used for measuring PM_{10} and $PM_{2.5}$ concentrations continuously. The PM monitors were calibrated every week using reference gauges with precisely-defined surface density. Relative humidity of the sample air was controlled to less than 35% using a Smart Inlet Heater system. Not only PM concentrations meteorology parameters (wind speed, wind direction, temperature, relative humidity, etc) were also recorded. Hourly PM concentration data was scrutinized to remove the invalid ones which were identified in several ways, (i) the ratio of $PM_{2.5}$ to PM_{10} must not be greater than 1.0, (ii) the ratio should not be unusually high or low compared to its neighboring ones; such ratio indicates error in instrument signal or unusual source encroachment, (iii) the data should not be fixed on a certain value for more than 2 consecutive hours. Year-wise rate of valid data at the stations are provided in Table 2.

2. 3 Conditional Bivariate Probability Function (CBPF)

Conditional Probability Function (CPF) shows the probability of a receptor to acquire from a particular wind direction sector a species greater than some speci-

Table 2. Year-wise rate (in percent) of valid data generated at the stations.

Station	Component	2013	2014	2015	2016	2017	2018 (up to April)
Dhaka	PM_{10}	93.0	89.3	73.1	75.0	90.6	90.6
	$PM_{2.5}$	91.1	91.3	93.0	68.7	93.0	92.2
Chattogram	PM_{10}	85.8	61.1	67.3	77.8	74.3	61.5
	$PM_{2.5}$	83.8	74.1	70.3	72.9	38.5	no data
Gazipur	PM_{10}	77.3	91.7	83.58	70.0	69.0	75.6
	$PM_{2.5}$	76.7	85.9	69.5	75.0	64.7	73.7
Narayanganj	PM_{10}	91.2	82.7	86.3	79.3	78.7	82.5
	$PM_{2.5}$	69.2	68.2	56.8	69.6	62.7	74.8
Sylhet	PM_{10}	81.8	87.3	64.6	66.4	84.5	83.0
	$PM_{2.5}$	82.1	86.2	66.2	63.3	82.2	79.7
Barisal	PM_{10}	89.3	59.6	80.0	70.2	22.2	82.3
	$PM_{2.5}$	85.1	85.6	80.7	77.0	61.6	no data

fied concentration value (usually 90 percentile). CPF is primarily used to find the directions having high probability of associating high concentrations of a pollutant at a site. However, the CPF can be usefully applied to the bivariate polar plots. In this case, the CPF along with a third variable (most of the cases wind speed, or any other meteorology variable) is plotted with the bivariate polar plots. The CPF in bivariate polar system which is termed as the conditional bivariate probability function (CBPF) can be defined as equation 1 (Uria-Tellaetxe and Carslaw, 2014):

$$CBPF_{\Delta\theta,\Delta u}=\frac{m_{\Delta\theta,\Delta u}(C\geq x)}{n_{\Delta\theta,\Delta u}} \quad \text{(Eq. 1)}$$

Where, $m_{\Delta\theta,\Delta u}$ is the number of samples in the wind sector $\Delta\theta$ with wind speed interval Δu having concentration C greater than a threshold value x, $n_{\Delta\theta,\Delta u}$ is the total number of samples in that wind direction-speed interval. *CBPF* can also be calculated for an interval of concentration rather than only values greater than some threshold. In that case, *CBPF* is equated as equation 2,

$$CBPF_{\Delta\theta,\Delta u}=\frac{m_{\Delta\theta,\Delta u}(y\geq C\geq x)}{n_{\Delta\theta,\Delta u}} \quad \text{(Eq. 2)}$$

Where, $m_{\Delta\theta,\Delta u}$ is the number of samples in the wind sector $\Delta\theta$ with wind speed interval Δu having concentration C between the value of x and y, $n_{\Delta\theta,\Delta u}$ is the total number of samples in that wind direction-speed interval (Uria-Tellaetxe and Carslaw, 2014).

2.4 Meteorology of the Cities

The meteorology of Bangladesh varies greatly in different seasons. Based on wind and rain pattern the seasons could be primarily divided into two, (a) dry season (November to April) and (b) wet season (May to October). For air quality study, Rana *et al.* (2016a) divides the dry season as (i) winter (November–January), and (ii) summer (February to April). Based on the data from the Department of Meteorology, Rana *et al.* (2016a) showed that the atmosphere as a whole in the country remains very dry during the month of February to April and very wet from June to August. The wintertime is characterized with low temperature, wind speed and mixing height while the summertime is typified with high temperature, dryness and mixing height, and clear sky (Rana *et al.*, 2016a). The meteorology also differs regionally; the northeast region of the country experiences more rainfall (annually $>$4000 mm) while the northwest region gets the less (annually $<$2300 mm) (Shahid, 2010). The climate of the northwest region is extreme (colder in winter and hotter in summer and wet season) while that of the northeast, south and southeast regions is moderate.

Four stations (Gazipur, Chattogram, Sylhel and Barisal) out of the six under this study have onsite meteorology monitors; the wind roses of these stations are shown in Fig. 2. Meteorological parameters were measured every minute, and an hourly average of a parameter was deducted from an arithmetic mean of the values in that hour. Measuring height of the parameters at the stations was about 15 meter.

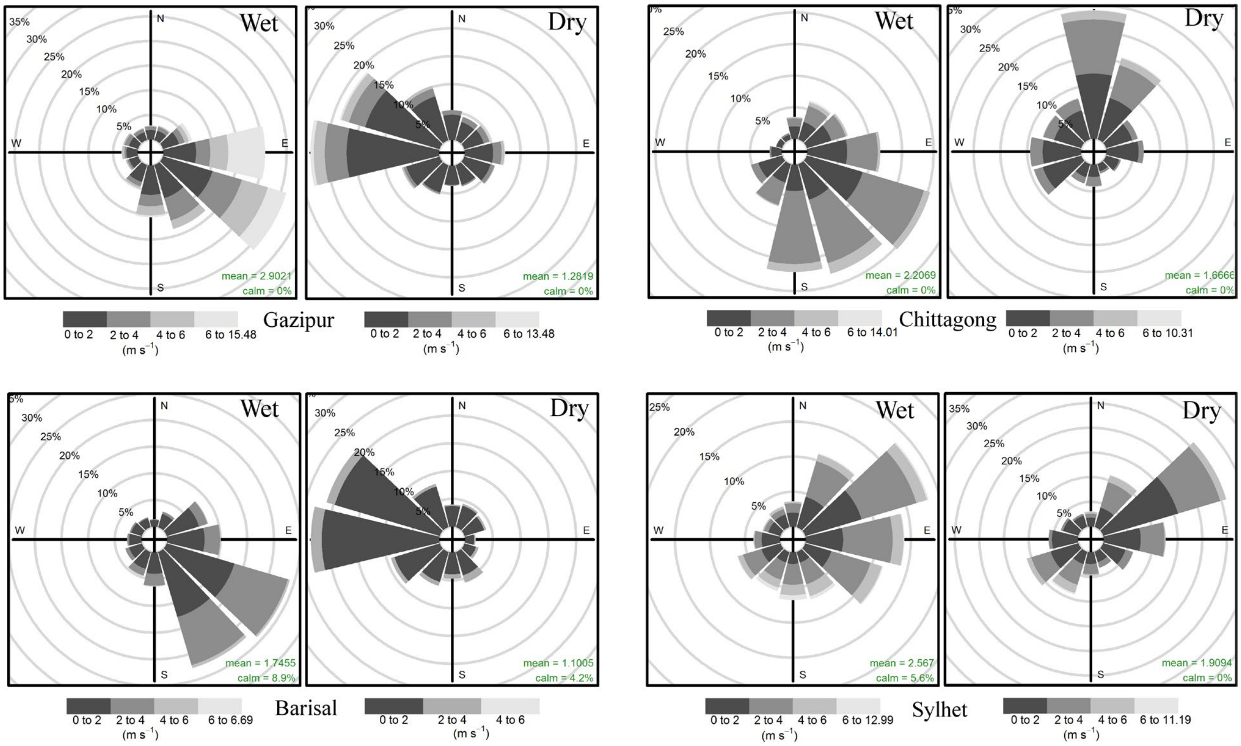

Fig. 2. Wind-roses at different stations in wet and dry season.

The winter and wet seasonal winds are opposite; the wind in general flows from the west and northwest directions during wintertime and from the south and southeast directions during summertime and wet season (Rana *et al.*, 2016a). This study found most of the regions except the northeast (Sylhet) and southeast (Chattogram) parts of the country followed this wind pattern (Fig. 2). Sylhet experienced major wind from the northeast direction in dry season and from the east, northeast, and southeast directions in wet season (Fig. 2). Dry seasonal wind in Chattogram was observed from the north and northeast directions, and the wet seasonal wind from the south and southeast directions.

3. RESULTS AND DISCUSSION

3.1 Overview of PM Concentrations

PM concentrations all over the country was found to follow a unique pattern of seasonal variations - high concentrations in dry season (November–April) and low in wet season (May–October) (Fig. 3). The contributions of $PM_{2.5}$ to PM_{10} were comparatively higher in wintertime (December–January) and lower in summertime (February–April). The contributions in wintertime in Dhaka, Chattogram, Narayanganj, Gazipur, Sylhet and Barisal cities were respectively 67.0, 64.0, 60.0, 70.0, 58.0, and 76.0% whereas the contributions in summertime were respectively 56.0, 53.0, 46.0, 58.0, 53.0 and 64.0%. $PM_{2.5}$ contributions to PM_{10} in wet season in the cities were 52.0, 55.0, 37.0, 58.0, 42.0 and 72.0% respectively. The driving forces for higher $PM_{2.5}$ contribution to PM_{10} in winter season could be the meteorology, open burning and the distant sources. Scare rainfall, low wind speed and low mixing height (Rana *et al.*, 2016a) in winter season aid in build up particulate concentrations in the atmosphere of this region. Compared to coarse particles, fine particles have high residence time in the atmosphere, and are expected to gain much over the coarse particles in dry atmosphere. Lots of seasonal sources especially brick manufacturing kilns, open stubble and garbage burning, etc. become active in this season and contribute greatly to particulate pollution. The meteorology of winter season in this region also assists in long range transportation of fine particulate matters. Azkar *et al.* (2012) investigated influences of long range

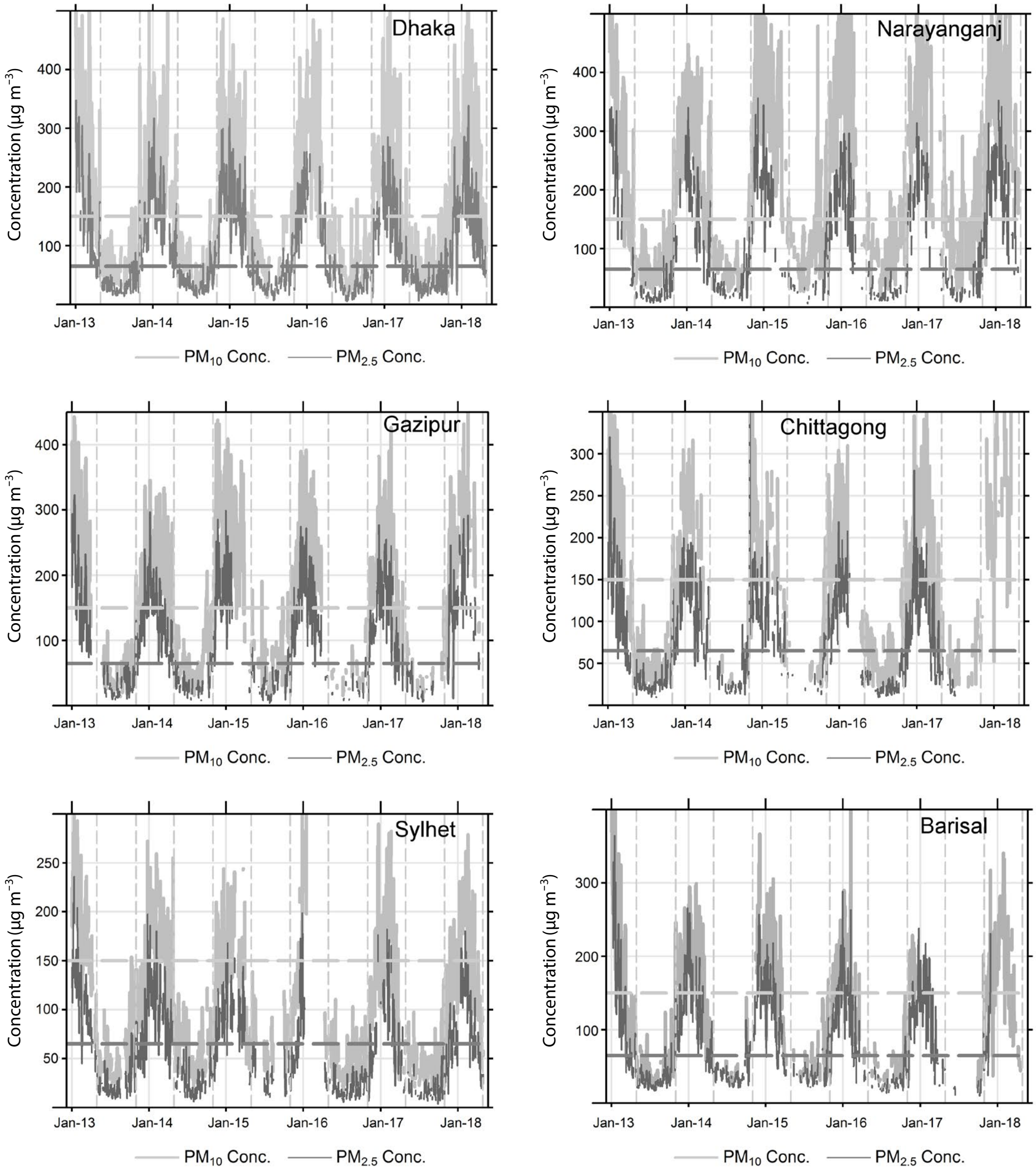

Fig. 3. Time series graph of 24-h average PM_{10} and $PM_{2.5}$ Concentrations at different cities of Bangladesh; horizontal lines are national limit values of PM_{10} (upper) and $PM_{2.5}$ concentrations (lower).

transport and brick kiln emissions on air quality over Bangladesh by using air quality simulations. Fine particles ($PM_{2.5}$) have higher health impact potential, and are related with complex respiratory and cardiovascular problems and may even create cancer in human being (Feng *et al.*, 2016). Thus, the air pollution during winter season in Bangladesh could be considered more damaging; extra care and management is needed to control the air pollution in this season. In contrast to the winter season, the wet season experiences fresher air for the frequent rainfall, high mixing height, etc.

The cities located in the middle of the country (Dhaka, Narayanganj, Gazipur) were found more polluted compared to other regions of the country, and that Narayanganj was found as the most polluted city in the country. Dhaka and Gazipur experienced almost the similar pollution level, although the Dhaka station is located at congested urban site and is influenced by heavy traffic, and the Gazipur site is sub-urban background with rare nearby sources. The Narayanganj station although received

Table 3. Overview of the daily PM concentrations in different seasons in the cities of Bangladesh. Daily concentrations were calculated with minimum 80% data availability.

City	Season	PM_{10} conc. ($\mu g\ m^{-3}$) Percentiles				$PM_{2.5}$ conc. ($\mu g\ m^{-3}$) Percentiles			
		25	50	75	Mean ± sd	25	50	75	Mean ± sd
Dhaka	Winter	222.7	278.7	342.7	288.0 ± 151.5	153.4	184.0	226.0	190.0 ± 93.0
	Summer	128.6	192.0	273.9	208.6 ± 152.0	65.7	97.8	147.6	110.0 ± 85.0
	Wet	48.5	62.6	88.4	72.5 ± 51.6	23.3	32.3	45.9	37.3 ± 27.3
	Annual*	66.3	114.8	229.3	155.8 ± 140.2	33.1	55.0	129.0	86.1 ± 85.0
Gazipur	Winter	210.6	256.0	310.6	262.0 ± 145.4	145.0	173.0	221.4	180.0 ± 96.0
	Summer	165.7	231.0	282.0	224.0 ± 132.2	83.5	121.1	157.6	122.3 ± 76.1
	Wet	33.4	48.7	77.9	60.3 ± 48.5	18.2	27.5	43.2	32.3 ± 26.1
	Annual	59.3	153.0	244.0	161.3 ± 136.0	34.8	88.8	155.4	100.7 ± 86.8
Narayanganj	Winter	317.0	377.0	432.0	377.0 ± 157.5	188.4	225.4	265.7	227.0 ± 102
	Summer	156.7	250.6	349.4	261.0 ± 156.4	68.5	125.0	187.8	132.3 ± 93.2
	Wet	55.7	83.7	120.0	99.2 ± 84.4	17.2	25.0	38.0	34.4 ± 34.0
	Annual	86.7	160.2	310.8	203.0 ± 163.2	26.0	57.4	173.0	108.0 ± 103
Chattogram	Winter	182.0	216.0	260.0	224.0 ± 113.0	116.6	138.5	167.8	143.3 ± 76.0
	Summer	108.0	166.4	237.6	173.8 ± 100.0	52.4	85.9	125.3	91.2 ± 63.4
	Wet	34.4	45.7	61.6	53.7 ± 40.8	17.6	22.3	31.3	28.4 ± 21.3
	Annual	51.8	113.7	200.2	132.2 ± 106.0	26.8	64.4	120.2	78.7 ± 106.0
Sylhet	Winter	135.7	170.3	199.4	172.7 ± 114.0	78.7	103.8	128.2	105.0 ± 76.0
	Summer	101.7	160	192.5	151.0 ± 92.0	56.8	84.0	110.5	83.4 ± 55.0
	Wet	34.4	47.5	64.1	52.0 ± 38.3	13.5	19.5	28.7	22.7 ± 21.1
	Annual	47.5	83.1	149.1	102.5 ± 91.2	19.8	38.8	83.6	54.6 ± 57.3
Barisal	Winter	184.0	210.4	245.6	218.4 ± 112.0	134.6	154.0	187.0	163.7 ± 82.3
	Summer	87.0	129.6	190.6	145.0 ± 97.0	52.0	83.5	124.5	92.7 ± 64.0
	Wet	34.0	44.6	63.0	56.0 ± 41.6	25.0	32.3	41.9	37.6 ± 24.3
	Annual	50.9	112.7	190.6	126.7 ± 102.7	34.0	66.7	130.7	85.6 ± 71.5

*Annual mean was calculated with the data from 2013 to 2017.

the highest level of both PM_{10} and $PM_{2.5}$ concentrations (Table 3), it had comparatively lower $PM_{2.5}$ to PM_{10} ratio-indicating massive sources of coarse particles near the station. PM concentrations captured at Gazipur site especially during dry season were too high (Table 3) for a sub-urban background station and comparatively higher $PM_{2.5}$ to PM_{10} ratio indicates effects of industrial activities and open cooking. People of Gazipur are used to burn logs, straws and leaves for cooking, and both Gazipur and Narayanganj cities are highly industrialized. Number of battery manufacturing industries, steel mills, dying factories, garment manufacturing units, pharmaceuticals, cement industries, etc. operate within the boundary of those two districts. $PM_{2.5}$ concentrations in Barisal city were found greater than that of Chattogram city which is bigger, busier, and more industrialized than Barisal city. Contributions of $PM_{2.5}$ to PM_{10} in wet season were found exceptionally high (0.72) in Barisal city. Barisal has a big river port and the diesel driven water vessels could be a possible source of higher PM pollution in the city. Among the cities, Sylhet was found the least polluted in respect of both PM fractions and fine particle contributions to PM.

The statistics provided in Table 3 were calculated from the daily averaged PM_{10} and $PM_{2.5}$ concentrations captured at the stations from 2013 to April 2018-each data thus representing individual day. Most of the days (~90%) of the winter season, especially during the month of December and January, the concentrations of both fractions of PM in the cities (except Sylhet) were greater than the national standards; in case of Sylhet, it is about 75% days of December and January when PM levels were noncompliant in respect to the Bangladesh National Standards (150 $\mu g\ m^{-3}$ for daily PM_{10} and 65 $\mu g\ m^{-3}$

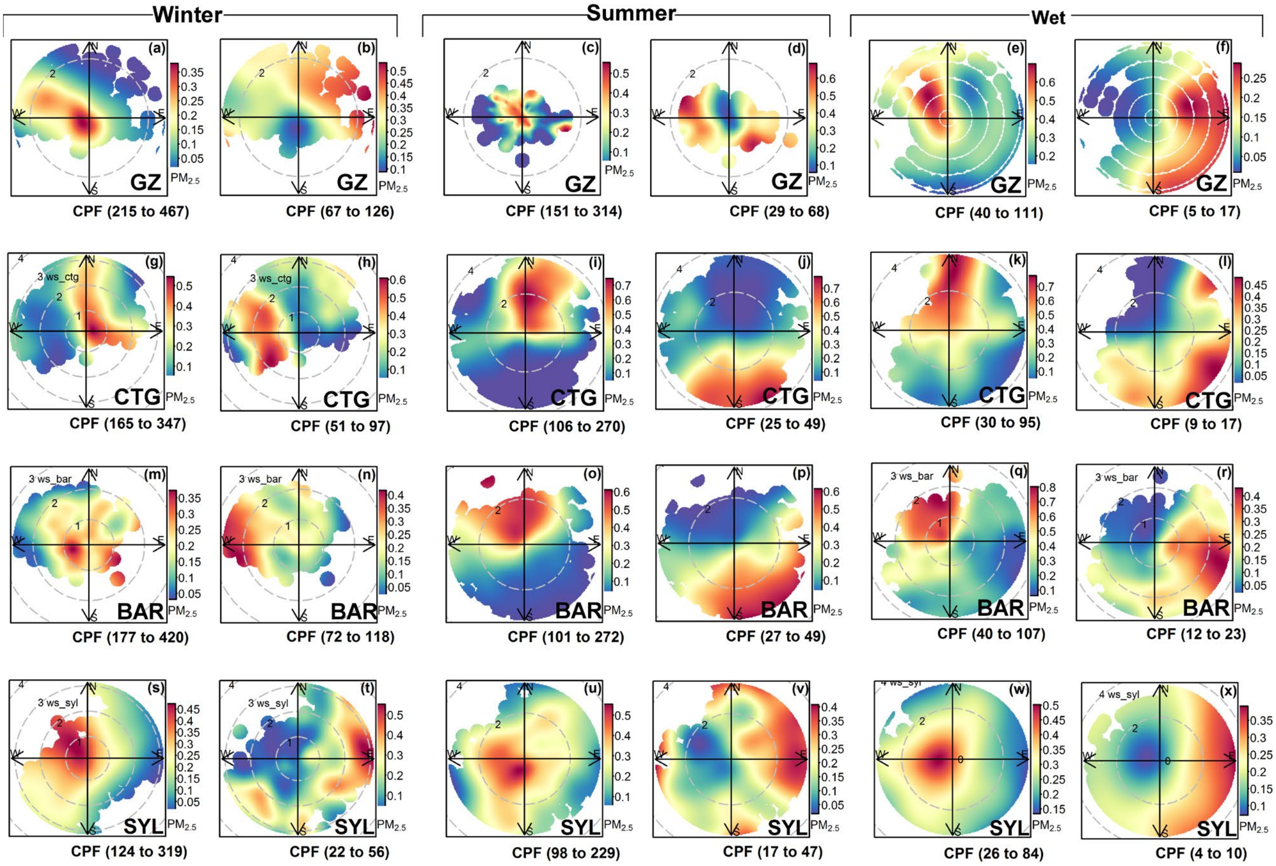

Fig. 4. CBPFs of $PM_{2.5}$ concentrations in two percentile ranges (70–98 and 5–30) shown for different seasons in the cities. GZ = Gazipur, CTG = Chattogram, BAR = Barisal and SYL = Sylhet. The color bar shows the CPF probability of $PM_{2.5}$ for the concentration range given in the parentheses below the figure.

for daily $PM_{2.5}$ concentrations). Compared to winter seasons, the summer seasons were found less polluted (Table 3); however, coarse particles were found to dominate over the fine particles in summer in contrast to the winter when fine particles dominated over the coarse particles (Table 3). On an average, the concentrations of PM_{10} and $PM_{2.5}$ in summer in Dhaka were respectively 27.7 and 41.0% lower than those in winter season; in other cities the reductions in PM_{10} and $PM_{2.5}$ concentrations from winter to summer were respectively as follows, Narayanganj 30.7 and 41.7%, Gazipur 14.5 and 32.2%, Chattogram 22.7 and 36.3%, Sylhet 12.5 and 20.5% and Barisal 33.6 and 43.4%. In all of the cities, decreases in $PM_{2.5}$ concentrations from winter to summer were greater than the reductions in PM_{10} concentrations from winter to summer.

The wet seasonal PM_{10} and $PM_{2.5}$ concentrations in the cities were very compliant with the NAAQS but still violated the WHO guideline values respectively in Dhaka by 75 & 75% days, Narayanganj by 75 & 50% days, Gazipur by 50 & 50% days, Chattogram by 50 & 50% days, Sylhet by 50 & 25% days, and Barisal by 50 & 50% days of the wet season (Table 3).

3.2 Directional Influences on the PM Concentrations at the Stations

CPF polar plots of $PM_{2.5}$ concentrations at low (5–30) and high (70–98) percentile intervals were examined at the stations particularly which had on-site meteorology data.

The CPF polar plots of $PM_{2.5}$ concentrations (Fig. 4) show that all of the stations were experiencing high level of $PM_{2.5}$ concentrations at calm weather during the winter season (Fig. 4a, g, m, s)-indicating most of the $PM_{2.5}$ being generated from the local sources. The Gazipur station during all of the seasons was affected severely with high level of $PM_{2.5}$ from the sources lying to the northwest and southwest directions (Fig. 4a, c, e); sources to the southeast directions were also crucial for high $PM_{2.5}$ concentrations especially during summer season (Fig. 4c). High $PM_{2.5}$ concentrations during all of the seasons at the Chattogram station were found associated with the eastern and northern winds (Fig. 4g, i, k); the impacts on this station from the south, northwest and southwest directions were comparatively weak (Fig. 4h, j, l). Barisal station was experiencing high $PM_{2.5}$ pollution from the west in winter season (Fig. 4m) and from the north during summer and wet seasons (Fig. 4o, q). The southern and southeastern sources were not contributing highly in any season in Barisal station (Fig. 4p, r). Sylhet station also observed high level of $PM_{2.5}$ from the northwest direction during winter season, and from the west during summer and wet seasons (Fig. 4u, w).

3.3 Diurnal Variations of PM Concentrations

The diurnal variations of PM concentrations (both fractions) in Dhaka, Narayanganj, Gazipur and Barisal were mostly the same in winter and summer seasons (Fig. 5)-the trends were bimodal in pattern, having peaks at 9:00 am and 9:00 pm. After the concentration peak at 9:00 am, PM concentrations (both fractions) in all of those cities steeply went down throughout the day; the concentrations started to rise from 6:00 pm till the peak at 9:00 pm. The concentrations plummeted throughout the night and started to rise again from 6:00 am next morning (Fig. 5). The wet seasonal diurnal trends of PM in those cities also experienced two pikes at 9:00 am and 9:00 pm, but the daytime variations of PM in this season were little different from the winter and summer seasons - tiny upward trend in the afternoon of wet season was noticed, and that the daytime trends of PM concentrations were little flat compared to those in winter and summer (Fig. 5). Diurnal variations of PM in the city of Chattogram and Sylhet were found little different from other cities discussed above, especially the PM peak in the morning in these two cities were less pronounced.

Although the cities of Chattogram, Sylhet and Barisal are located about 300 km away from the middle part of the country (Dhaka, Narayanganj, Gazipur) and have different type of responsible sources of PM, the diurnal variations of the PM concentrations in all of the cities especially after 6:00 pm were the same (Fig. 5). This is perhaps for the similar trend characteristics in the mete-

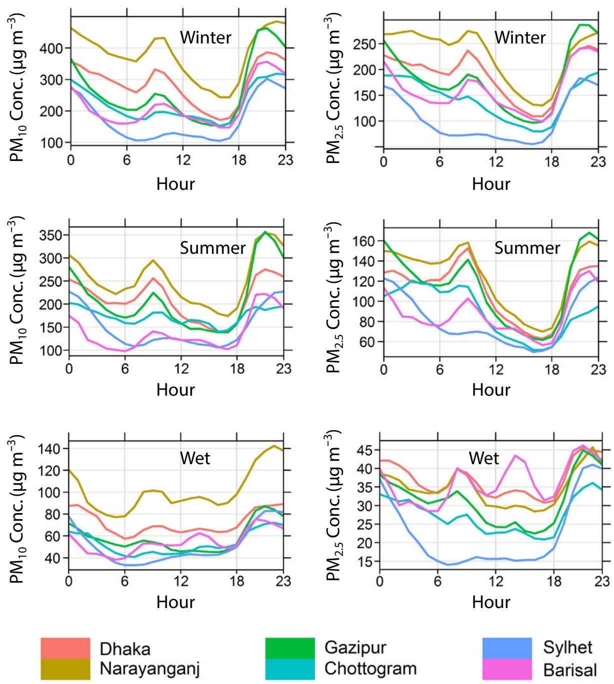

Fig. 5. Diurnal trends in PM_{10} and $PM_{2.5}$ concentrations in different seasons in the cities.

orology parameters in the cities (Fig. 6). The country of Bangladesh (except Sylhet and Chottogram regions) has a very flat terrain; climatic changes throughout the country are almost similar. Correlative trends of normalized $PM_{2.5}$ concentrations and meteorological parameters are shown in Fig. 6.

Meteorology has profound impact on the air quality in a region (Schnell *et al.*, 2018; Tiwari *et al.*, 2014). Collective impacts of the local emissions, meteorology parameters, and boundary layer dynamics determine the trends in the PM concentrations in a region (Dumka *et al.*, 2015, 2013). Stull (1988) suggests that the fumigation effect and the evolution of the boundary layer height (BLH) just after the sunrise favor building up PM concentrations, which when combined with the rush hour emissions from especially motor vehicles result in the morning peaks. As Chattogram and Sylhet stations were far from the busy roads, and any other big local sources, the morning peaks of PM concentrations at those stations could not develop much (Fig. 5). However, nighttime hike in PM concentrations at 9:00 pm was very common in all of the seasons in the cities (Fig. 5).

Although the source activities (vehicles, road dust, constructions, industries, etc.) in the cities run in large

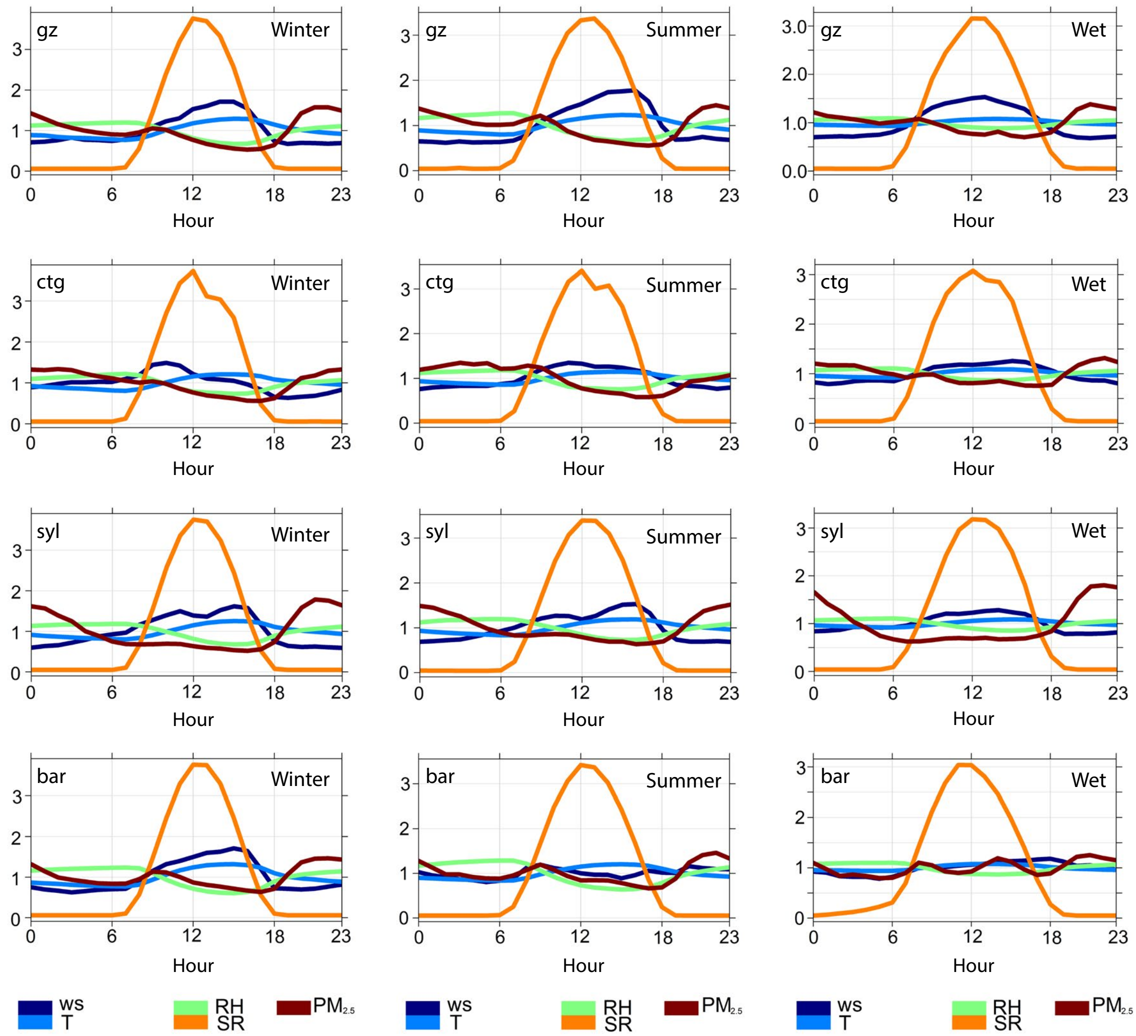

Fig. 6. Relationships among normalized level of diurnal variations of $PM_{2.5}$ and meteorology parameters; ws = wind speed, T = temperature, RH = relative humidity, SR = solar radiation. gz = Gazipur, ctg = Chattogram, syl = Sylhet, bar = Barisal.

scale at daytime, the PM concentrations were found to increase at nighttime (Figs. 5 & 6) and the average nighttime PM concentrations in all of the cities were found greater than the average daytime concentrations (Table 4). Fig. 6 reveals that the daytime meteorology of the cities was characterized with comparatively greater solar radiation, wind speed and temperature, and lower relative humidity. Nighttime PM_{10} and $PM_{2.5}$ concentrations at Dhaka stations in winter season were on average 33.5 and 26.4% greater than the daytime concentrations respectively. Similarly, the nighttime PM_{10} and $PM_{2.5}$ concentrations were greater than the daytime concentrations in other cities in winter respectively by 26.2 and 22.0% in Narayanganj, 63.8 and 54.6% in Gazipur, 45.6 and 45.5% in Chattogram, 85.4 and 98.7% in Sylhet, and 38.2 and 35.2% in Barisal. On the other hand, the increase in the nighttime PM_{10} and $PM_{2.5}$ concentrations compared to daytime concentrations in summertime in the cities were 23.2 and 16.7% in Dhaka, 22.8 and 23.7% in Narayanganj, 46.8 and 38.8% in Gazipur, 13.6 and 21.5% in Chattogram, 52.3 and 57.6% in Sylhet, and 28.6 and 24.2% in Barisal. Thus, the increase in nighttime PM_{10} concentrations compared to that in $PM_{2.5}$ concentrations during both winter and summer seasons were found higher in Dhaka, Gazipur and Barisal. Narayanganj experienced mostly similar increases in both fractions of PM concentrations at nighttime; however, Chattogram and Sylhel experienced higher increase in $PM_{2.5}$

Table 4. Average PM concentrations (μg m^{-3}) at daytime and nighttime in different seasons in the cities.

		Winter		Summer		Wet	
		Day (avg ± sd)	Night (avg ± sd)	Day (avg ± sd)	Night (avg ± sd)	Day (avg ± sd)	Night (avg ± sd)
DHK	PM_{10}	243.4 ± 122.0	325.6 ± 162.2	186.3 ± 131.3	229.6 ± 164.2	64.7 ± 38.7	78.5 ± 60.0
	$PM_{2.5}$	166.0 ± 84.0	209.8 ± 95.4	100.8 ± 80.4	117.6 ± 87.5	34.1 ± 23.1	39.5 ± 30.2
NG	PM_{10}	331.3 ± 143.8	418.2 ± 155.5	224.5 ± 127.5	275.8 ± 173.3	93.0 ± 66.2	107.0 ± 96.5
	$PM_{2.5}$	203.9 ± 105.0	248.6 ± 96.0	110.0 ± 85.2	136.1 ± 97.7	32.1 ± 29.0	37.0 ± 39.2
GZ	PM_{10}	195.8 ± 81.0	320.7 ± 159.2	167.1 ± 82.1	245.4 ± 153.2	49.4 ± 36.2	66.2 ± 55.6
	$PM_{2.5}$	139.0 ± 66.7	214.9 ± 101.2	94.4 ± 61.5	131.0 ± 82.7	27.1 ± 22.3	35.5 ± 28.1
CTG	PM_{10}	178.8 ± 71.7	260.3 ± 127.0	162.0 ± 82.2	184.0 ± 111.5	46.0 ± 26.0	59.6 ± 49.3
	$PM_{2.5}$	114.8 ± 59.4	167.0 ± 81.6	80.8 ± 57.7	98.2 ± 67.0	23.8 ± 15.2	30.8 ± 25.2
SYL	PM_{10}	118.3 ± 51.5	219.4 ± 128.6	116.5 ± 54.0	177.4 ± 106.0	40.2 ± 20.0	60.8 ± 46.7
	$PM_{2.5}$	68.0 ± 41.7	135.1 ± 83.0	61.8 ± 35.8	97.4 ± 62.3	15.5 ± 11.2	28.5 ± 25.2
BAR	PM_{10}	180.2 ± 58.6	249.0 ± 133.4	118.0 ± 57.0	151.8 ± 118.0	52.1 ± 30.7	55.6 ± 48.7
	$PM_{2.5}$	135.0 ± 49.8	182.5 ± 96.4	76.3 ± 44.5	94.8 ± 74.8	35.5 ± 20.0	36.1 ± 27.1

DHK = Dhaka, NG = Narayanganj, GZ = Gazipur, CTG = Chittagong, SYL = Sylhet, BAR = Barisal

concentrations compared to that of PM_{10} concentrations at nighttime during both winter and summer seasons. Coarse particles ($PM_{2.5\text{-}10}$) have comparatively shorter residence time in the atmosphere compared to fine particles (Seinfeld and Pandis, 1998); the fine particles are usually expected to dominate at nighttime when major source activities are ceased off. The increase in PM_{10} concentrations compared to $PM_{2.5}$ concentrations at nighttime indicates presence of some sort of source activities at nighttime in Dhaka, Gazipur and Barisal cities. Chattogram and Sylhet stations are located far from major local sources and thereby experienced relatively higher $PM_{2.5}$ concentrations at nighttime.

3.4 Long Range PM Sources

In dry weather PM gets longer residence in the atmosphere, especially the fine particles may have weeks of residence time and travel hundreds to thousands of kilometers before being removed from the atmosphere by dry deposition (Seinfeld and Pandis, 1998). The entire south Asian region during the dry season suffers from increased level of PM in air. Industries, soil dust open cooking and stubble burning, etc. altogether emit tons of PM into the atmosphere of south Asian countries. The PM so emitted during dry season disperses around thousands of kilometer over the south Asian countries (Begum *et al.*, 2011; Lawrence and Lelieveld, 2010). Several researches earlier have shown incursions of trans-boundary PM into Dhaka, and the source locations identified by those studies are Nepal and the Indian regions lying on the Himalayan Valley and on the Indo-Gangetic Plain (Rana *et al.*, 2016b; Begum *et al.*, 2010). In all of the previous works, trans-boundary pollution was investigated over Dhaka and its surroundings; however, in this paper, long range PM pollutions (both within and beyond the national boundary) were investigated around Dhaka, Chattogram, Sylhet and Barisal cities.

Ninety six (96)-hour backward isobaric trajectories with every 3-hours interval during the period of 15 December 2015 to 15 January 2016 on Gazipur, Chattogram, Sylhet and Barisal stations were calculated using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT-4) model (Draxler and Rolph, 2003) on the platform of the analytical software "R" which was used to analyze the trajectories and concentration data. Global Reanalysis Meteorology Data downloaded from the archive of National Oceanic and Atmospheric Administration (NOAA) was used as input to the model. As those trajectories were analyzed with their association with station concentration data, starting points of the trajectories were kept at 10.0 m high from the ground. PM concentrations in the cities usually remain at the highest level during this time period (Fig. 3). Hourly $PM_{2.5}$ concentrations at the stations were associated with the trajectories arrived at the respective hours at the stations. Concentration weighted trajectory (CWT) method (seibert *et al.*, 1994) was applied to identify possible long

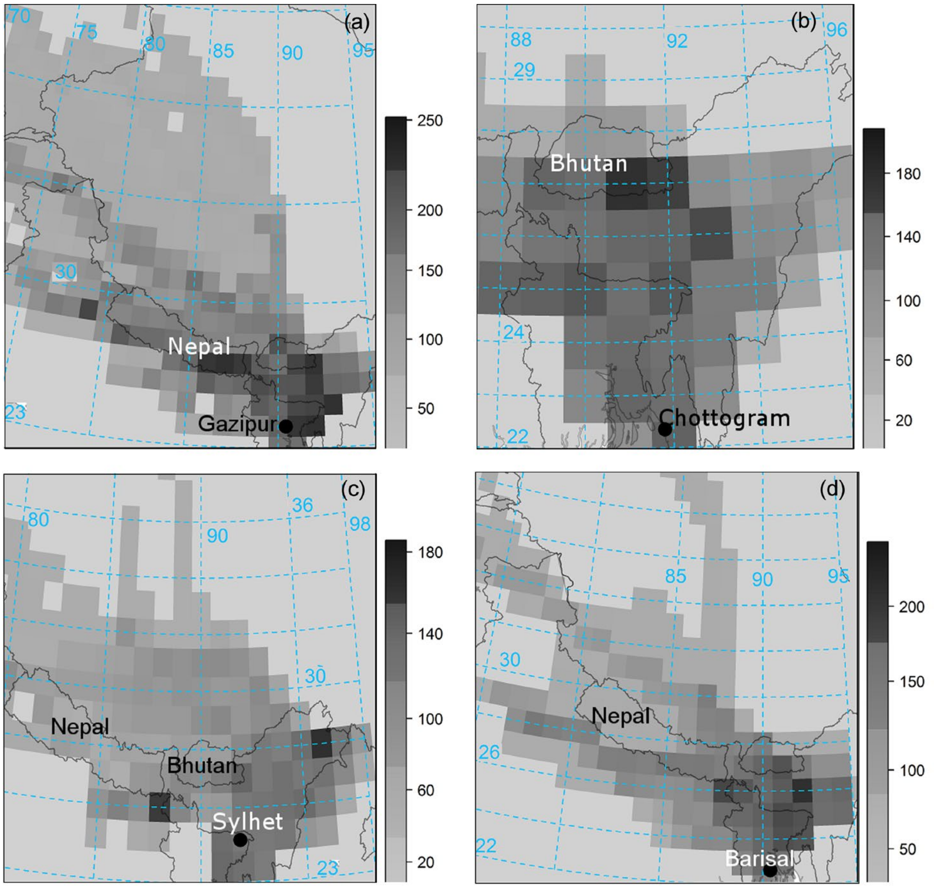

Fig. 7. Gridded back trajectory concentrations showing mean $PM_{2.5}$ concentrations on each grid cell of 1° × 1° area, using CWT method.

range regions responsible for contributing to PM pollution at the stations. CWT is calculated from the residence time of trajectories over a grid cell and the respective $PM_{2.5}$ concentrations the trajectories were associated with. In this method, for each cell of a domain, mean CWT or logarithmic mean concentration of a pollutant species was calculated according to the Equation 3 as follows:

$$\ln(\overline{C}_{ij}) = \frac{1}{\sum_{k=1}^{n}\tau_{ijk}} \sum_{k=1}^{n} \ln(C_k)\tau_{ijk} \qquad \text{(Eq. 3)}$$

where i and j are the indices of grid, k the index of the trajectory, n the total number of trajectories used in analysis, C_k the pollutant concentration ($PM_{2.5}$ conc. in this case) measured upon arrival of trajectory k, and τ_{ijk} the residence time of trajectory k in grid cell (i, j). A high value of $\overline{C}_{ij}$ means that, air parcels passing over cell (i, j) would, on average, cause high concentrations at the receptor sites.

Gridded trajectory concentrations in Fig. 7 show that the stations were possibly receiving $PM_{2.5}$ emitted from the long range sources both within and outside the national boundary. Within the boundary, the sources in the northern districts of Naogaon, Bogura and Rangpur were contributing to fine particles in the middle (Fig. 7a), south (Fig. 7d), and southeast (Fig. 7b) parts of the country. The south (Fig. 7d) and southeast (Fig. 7b) regions were also influenced by the sources in the middle of the country (Dhaka, Gazipur, and Narayanganj). Sources in the eastern districts (Narsindhi and Bramhanbaria) and northeastern districts (Sylhet) were also found contributing to $PM_{2.5}$ concentrations at the south (Fig. 7d) and southeast (Fig. 7b) stations. Sylhet station was found gaining $PM_{2.5}$ from the sources located in the middle of the country (Fig. 7c) during the time from December 2015 to January 2016. The northern districts possess a large number of rice mills and brick kilns; rice husks are widely used as fuel in those rice mills, and the brick kilns burn coals. Agricultural residue burning and open cooking are also broadly practiced in the northern districts. Emissions from these sources may have contributed to $PM_{2.5}$ concentrations in other parts of the country (Fig. 7).

Sources of Nepal, and Delhi-NCR and Uttar Pradesh regions of India could have contributed to the fine particles at the Gazipur station (Fig. 7a). Delhi-NCR is currently the top ranked polluted city in the world (WHO, 2018); Nepal and Pakistan are also heavily polluted (Gurung and Bell, 2012; Khatum, 2017). The stations could have also gained PM from the eastern state of Tripura and northeastern state of Assam/Meghalaya of India (Fig. 7). Barisal station was also found to have probability of getting PM from Nepal and its neighboring Indian regions (Fig. 7). Wind directions during dry season in Sylhet were different from those in Gazipur and Barisal (Fig. 2). While Gazipur and Barisal experienced air that entered Bangladesh territory through the western or northern borders, Sylhet received air that crossed the border through the north eastern borders. Thus, the Sylhet station was much affected by the sources in Meghalaya and Assam rather than sources in Nepal and its neighboring regions (Fig. 7). Sources of Meghalaya and Assam could also be crucial for PM pollution in other parts of Bangladesh as shown by Fig. 7.

4. CONCLUSIONS

Trend characteristics of PM concentrations in six cities of Bangladesh from 2013 to April 2018 were exhaustively studied. The cities were Dhaka, Gazipur and Narayanganj in the middle of the country, Chattogram to the southeast, Sylhet to the northeast and Barisal to the south of the country. The concentrations were monitored every minute with the continuous PM monitoring system BAM 1020 of the Met One Instrument Inc, USA. Two separate systems of the BAM 1020 were applied for monitoring the PM_{10} and $PM_{2.5}$ concentrations. After the study the following observations and findings were noted,

a) Atmospheric PM concentrations in all of the regions of the country were notably influenced by the seasonal variations. Average PM_{10} concentrations at Dhaka, Chattogram, Narayanganj, Gazipur, Sylhet and Barisal stations in dry seasons (November–April) were 238.7 ± 155.4, 190.7 ± 108.5, 303.6 ± 161.4, 227.3 ± 142.7, 151.7 ± 105.0 and 170.7 ± 108.4 μg m^{-3} respectively whereas those in wet seasons (May–October) were 75.0 ± 51.6, 55.5 ± 40.8, 102.4 ± 84.4, 60.6 ± 48.5, 52.7 ± 38.3, and 54.4 ± 41.6 μg m^{-3} respectively. The ratios of $PM_{2.5}$ concentrations to PM_{10} concentrations were comparatively higher in wintertime (December–January) and lower in summertime (February–April). The ratios in winter/summer time in Dhaka, Chattogram, Narayanganj, Gazipur, Sylhet and Barisal cities were 0.67/0.56, 0.64/0.53, 0.60/0.46, 0.70/0.58, 0.58/0.53 and 0.76/0.64 respectively. Ratios in wet season were 0.52, 0.55, 0.37, 0.58, 0.42 and 0.72 respectively.

b) The middle area of the country (Dhaka, Narayanganj and Gazipur) was found more polluted compared to other parts of the country, and the northeast region (Sylhet) was found the least polluted. Primary investigations reveal a relation between wind pattern and PM pollution level in the country-the northern, middle and southern part of the country (Dhaka, Narayanganj, Gazipur, Barisal) experienced wind mostly from the west and north-west directions during dry season and were characterized with very high PM pollution as well as higher contribution of fine fraction to the PM concentrations. In contrast, the southeast (Chattogram) and northeast (Sylhet) regions of the country having wind directions other than west and northwest directions in dry season experienced less pollution.

c) The diurnal variations of PM concentrations (both fractions) in Dhaka, Narayanganj, Gazipur and Barisal were mostly the same in winter and summer seasons-the variations were bimodal in pattern, having peaks at 9:00 am and 9:00 pm. Diurnal variations of PM in the city of Chottogram and Sylhet were found little different from other cities, especially the PM peak in the morning in these two cities were less pronounced. The nighttime PM_{10} and $PM_{2.5}$ concentrations at Dhaka stations in winter season were on average 33.5 and 26.4% greater than the daytime concentrations respectively. Similarly, the nighttime PM_{10} and $PM_{2.5}$ concentrations were greater from the daytime concentrations in other cities in winter respectively by 26.2 and 22.0% in Narayanganj, 63.8 and 54.6% in Gazipur, 45.6 and 45.5% in Chattogram, 85.4 and 98.7% in Sylhet, and 38.2 and 35.2% in Barisal. Daytime meteorology of the cities was characterized with comparatively greater solar radiation, wind speed and temperature, and lower relative humidity. Mixing height thus formed remains very deep at daytime compared to nighttime, giving a general tendency to generate high

PM concentration at nighttime.

d) The study found that the sources from one region (within the boundary of the country) were contributing to PM in other regions located downwind. For example, some hotspots in the northwestern districts (Naogaon, Bogura, Rangpur) were identified contributing to PM concentrations in the middle and south part of the country. Outside the boundary, sources in Nepal, and Delhi-NCR and Uttar Pradesh regions of India could have contributed to fine particles at the Gazipur station. The stations could have also gained PM from the eastern state of Tripura and northeastern state of Assam and Meghalaya of India.

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