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Assessment of acid deposition over Dhaka division using CAMx–MM5 modeling system

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ABSTRACT

This study provided an insight into the complex phenomenon of acid deposition over Dhaka division of Bangladesh. The 10– day HYSPLIT back trajectories showed that majority of air masses arriving to Bangladesh were originated from the west. The gridded emissions (0.055° or ~5.5 km) for Dhaka division was prepared for 2006 by updating a preliminary emission inventory (EI) available for 2000 and by conducting a new EI for two major source categories, brick kilns and traffic. In Dhaka division as of 2006, the brick kilns contributed the largest SO_2 emission (about 70%), while residential emissions had the highest share of CO and PM_{10} (over 60%) and substantial NMVOC (about 40%). Emission rates of SO_2 and NO_x in the dry season of 355 t/d and 183 t/d, respectively, were higher than the corresponding rates in the wet season of 60 t/d and 146 t/d, which was mainly due to the operation of brick kilns in the dry season. The acid deposition was simulated, using CAMx–MM5 model, for December and June to represent the dry and wet seasons, respectively. Model performance was reasonable considering the simulated spatial distribution of acid deposition with simulated wind and precipitation fields. The model results for SO_2 and NO_x concentrations were in the same ranges of the limited monitoring data available. However, the acid deposition simulation was still largely experimental due to the lack of acid deposition monitoring data for the model evaluation. The preliminary acid deposition simulation results suggested that the nitrogen wet deposition was the major contributor over the Dhaka division that could be the reason of exceeding the critical load for the local ecosystem. Further research is still required to refine the emission inventory and to gather the monitoring data to confirm the modeling results.

Keywords:
Emission inventory
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1. Introduction

Once released, air pollutants, such as SO_x and NO_x, are transported over long distances. During their atmospheric residence time, they can be transformed to form new pollutants. They may be deposited from the atmosphere as dry or wet particles or as vapors causing acidifying effects on the earth surface. This phenomenon is known as acid deposition, which incorporates both wet (with precipitation) and dry deposition to the earth surface (ISU, 2006).

The impact of acid deposition on an ecosystem depends on its assimilation capacity of the excess acidity, usually caused by NO_3^- and SO_4^{2-}, which in turn, depends on the components of the ecosystem (IIASA, 1994). The level of sensitivity of an area to acid deposition is usually indicated by biotic communities (Kuylenstierna and Chadwick, 1989). An ecosystem can tolerate the acid deposition as long as it is below the critical deposition load, i.e., the level that would not cause chemical changes leading to long–term harmful effects on essential ecosystem properties (Kamari, 1989).

Bangladesh has a population of 150 millions (BBS, 2008) and an agriculture based economy. Air pollution is a serious issue of concern in the country. Multiple sources, such as vehicles, industries, open biomass burning, etc., release a large amounts of particulate matter (PM) and toxic gases into the atmosphere. In addition, the country may also receive the Long–Range Transport (LRT) air pollution that is originated in upwind countries and arrived into the country following the prevalent monsoons. Thus, acid precursor emissions from both the local and regional sources may cause the acid deposition in the country (IIASA, 1994; Hicks et al., 2008). This may affect especially central part of Bangladesh where soil is of acidic nature (SRDI, 2008) and would harm the agricultural crops, the main product of the country. However, at present there is no adequate quantitative information on the potential acid deposition and potential effects for Bangladesh.

This study attempts to analyze the potential acid deposition threat to Bangladesh using an integrated approach that combines a source emission inventory (EI), air mass trajectory analysis, and three dimensional (3D) air quality modeling system: Comprehensive Air Quality Model with Extensions (CAMx) driven by Mesoscale meteorological Model (MM5) or CAMx–MM5. Due to the current lack of monitoring data, the modeling approach appeared to be a cost–effective way to assess the acid deposition over the study domain.

2. Methodology

2.1. Study area

Bangladesh is divided into six major divisions and 64 districts administratively. Dhaka division (Figure 1), located in the centre of Bangladesh, was selected as the study area. The division has an area of 31 120 km^2 consisting of 17 districts (Banglapedia, 2008) with population of about 39 millions, i.e. about 31% of the total population of the country (BBS, 2008).

Bangladesh has a tropical monsoon climate, withhot and rainy summers (April–September) and a dry winters (November to February). Mean annual temperature of the country is about 25 °C.

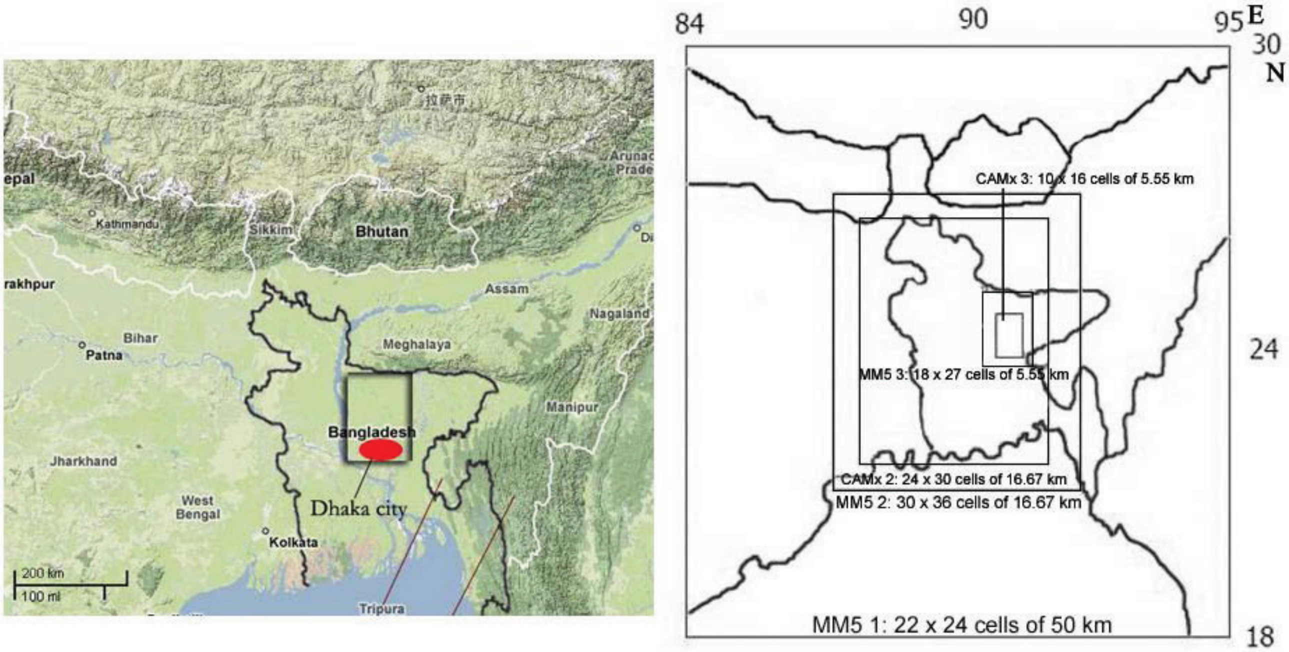

Figure 1. *Map of Bangladesh indicating its location in Asia, the modeling domain in Bangladesh and the CAMx–MM5 modeling domain structure (Dhaka city is indicated).*

April is the hottest month, when the mean temperature ranges from 27 °C in the south eastern to 31 °C in the northwestern part of the country with the country wise monthly mean of 30 °C. Average temperature in January, the coolest month, varies from 17 °C in the northwestern and northeastern parts of the country to 20 – 21 °C in the coastal areas with the country–wise mean of 18 °C. The annual minimum temperature is observed in late December or early January that may drop to as low as 3 to 4 °C in the extreme northwestern part. In the study area of Dhaka, the average January temperature is about 19 °C and the average April temperature is about 29 °C (Banglapedia, 2008; BMD, 2008).

Bangladesh is one of the wettest countries in the world. The average annual rainfall over the country is about 1 525 mm while in hilly areas the value may be as high as 5 080 mm. Most of rain occurs during the monsoon months (June–September) and little rain occurs in winter (November–February). Generally, winds are stronger in summer (ranging between 2.2 and 4.4 m/s) than in winter (ranging between 0.83 and 1.6 m/s) (Banglapedia, 2008; BMD, 2008). Figure 2 shows the meteorological conditions measured at a station in Dhaka for the year 2006.

The Dhaka division has numerous emission sources including traffic, industries of various scales ranked from traditional to advanced technologies, agricultural residue burning and residential combustion. In 2006, about 230 000 vehicles of different categories were running on the roads of the Dhaka division. Each month thousands of new vehicles were being added to the fleet. New industries are established regularly to meet the economic development, most of which are in Dhaka division to grab maximum facilities. In Dhaka division about 436 000 industries of different categories were in operation in 2006, which was increased 59% as compared to 2000 (BBS, 2008).

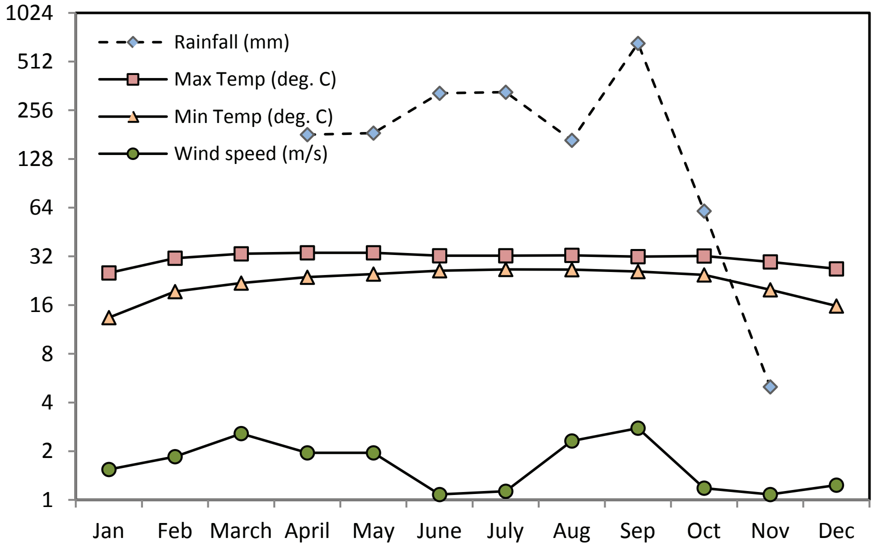

Figure 2. *Meteorological data of Dhaka station indicating monthly average maximum temperature; monthly average minimum temperature; monthly average rainfall and monthly average wind speed for 2006 (Source: BMD, 2008).*

2.2. Gridded emission inventory for model input

In this study we produced an emission inventory (EI) for 2006. For our CAMx modeling purpose, the emissions were calculated on the grid of 0.055° x 0.055° (about 5.55 km) for the Dhaka division.

There is a preliminary EI for the country which was prepared under the Male's Declaration project (Hasan, 2000) that presented annual average emissions for Bangladesh for the year 2000. We projected the existing EI of 2000 to 2006 using actual growth of selected sectors following the method described in Nghiem and Kim Oanh (2008). It was assumed that the technology levels in these sectors remain more or less same, thus only the growth was considered in the projection. The base year of 2006 was selected primarily because the emissions of the outer domains of our modeling studies was extracted from the CGRER, where the most recent data was for 2006.

In addition, a detailed emission inventory for 2006 was conducted for two major emission source categories: brick kilns and vehicles. Numerous polluting coal–fired brick kilns, a major source of SO_2 emissions, were not considered in the existing 2000 EI (Hasan, 2000) hence there was a need to include this source category. For the urban traffic in the study area, there was a shift from gasoline and diesel powered vehicles to compressed natural gas (CNG) starting in 2003 hence a new EI was required.

The EI was conducted for these two source categories following the general formula given in IPCC (1996) and elsewhere that are shown in Equation (S1) (see the Supporting Material, SM). The emission factors (EFs) for brick kilns used in this study (see the SM, Table S1) were the energy content based, kg/TJ, that were mainly selected from those provided by IPCC (1996) for coal and fuel wood. Thus, the emissions of the pollutants from soil in raw bricks during firing at high temperature were not considered. The EFs for NO_x, carbon monoxide (CO) and non–methane volatile organic compounds (NMVOC) for brick kilns were selected based on the fuel and kiln technology types. The EF of SO_x was calculated (Equation S2, see the SM) based on sulfur content of coal which commonly is 5% for the coal used in brick firing in the country (SOE, 2005; BMA, 2008). Note that no flue gas emission control is applied in brick kilns in Bangladesh. For PM_{10} the EF (kg/t) provided by US EPA (1999) for fired bricks was used.

The emissions of SO_x, NOx, CO, NMVOC and PM from the urban transport was estimated following the general formula given by IPCC (1996) and elsewhere (Equation S3, see the SM). The vehicle population and travelled distance (VTK) data for different broad types of vehicles (two–stroke and 4–stroke motorcycles, light duty and heavy duty diesel, light duty and heavy duty gasoline, CNG vehicles) were taken from Bangladesh Bureau of Statistics (BBS, 2008) that are also shown in Table S2 (see the SM). The vehicle EFs used (Table S3 see the SM) have been previously compiled for the country by the Bangladesh State of Environment report (SOE, 2005). Thus, other factors affecting the traffic emissions such as age distribution of the vehicle fleet or engine standards, e.g. EURO standards, were not considered in this study. In addition, the water transport sub–sector which likely has a large emission share was also excluded from our current EI. The ship emissions, in particular, may contribute more significantly to the emission over the whole country rather than to our study domain of the Dhaka division.

2.3. Long–range transport

The potential of LRT air pollution to Dhaka was assessed using the HYSPLIT trajectory modeling (Draxler and Hess, 1997). The starting point of the backward trajectory analysis was the Dhaka city of Bangladesh (lat: 23°43' N; lon: 90°24' E), 1 000 m above ground, and at 5:00 UTC (Coordinated Universal Time) or 11:00 (noon) Bangladesh local standard time (LST). The height was chosen to be 1 000 m, the average daytime mixing height for the Dhaka city (BMD, 2008), in order to minimize the friction effects of the earth surface (Pongkiatkul and Kim Oanh, 2007). The 10 day back trajectory was determined for each day of the year 2006. The clustering technique provided by the HYSPLIT 4.8 model was applied to identify the main trajectory patterns in the year as well as for selected periods of 3D simulation (June and December).

The meteorological data required for modeling was taken from the ARL (2008) data server (Gridded meteorological data archives) of the National Oceanic and Atmospheric Administration (http://www.arl.noaa.gov/ss/transport/archives.html). The meteorological input data, available 4 times a day (0, 6, 12 and 18 UTC), consisted of 3D winds, momentum and heat flux, relative humidity, temperature and pressure at the surface and 12 vertical layers (20, 50, 100, 150, 200, 250, 300, 400, 500, 700, 850, 925 and 1 000 hPa).

2.4. CAMx–MM5 modeling

Simulation of the acid deposition in Dhaka was done using the CAMx model (Environ, 2008) which was driven by MM5 modeling system (MM5, 2009). The chemistry mechanism used was the Carbon Bond version IV (CBIV). CAMx considers both dry and wet depositions and provides separate output of dry and wet deposition. The wet deposition considers precipitation parameters, scavenging of gases in cloud water, scavenging of ambient gases, solubility limit on gas scavenging, scavenging of aqueous aerosols and dry particles. In case of dry deposition, the model calculates the deposition velocities for each species and uses the values as the lower boundary conditions for vertical diffusion (Environ, 2008).

To generate the meteorological fields, MM5 model was run with a combination of different incorporated schemes. In this study we used Schultz scheme for explicit moisture content (IMPHYS), Rapid Radiative Transfer Model scheme (RRTM) for radiation (IFRAD), Grell scheme for Cumulus Parameterization (ICUPA), Medium Range Forecast scheme for planetary boundary layer and diffusion (IBLTYPE), Noah scheme for surface (ISOIL) and no convection was considered for shallow cumulus convection (ISHALLO).

Land use data for the modeling domain was taken from USGS (US Geological Survey) database (http://www.usgs.gov). The simulation was done for two months of the year 2006: June to represent the wet and December to represent the dry season. These two months were selected as they are at the middle of wet and dry season of the country. This was necessary as the computational system used was not sufficient for a longer simulation period within the scope of this study. The meteorological input data for MM5 were provided by the US National Center for Environmental Prediction (NCEP, 2009) 1° x 1° resolution final analysis (FNL), available online at (www.dss.ucar.edu/datasets/ds083.2/data./webgroups.html). The FNL data contain horizontal, vertical winds, temperature, specific humidity, cloud cover, geo–potential height, soil moisture, and soil temperature. The data is available for every six hours starting at 00 UTC. The surface meteorological data was collected from the Bangladesh Meteorological Department.

The structure of CAMx–MM5 system domains is given in Figure 1, which shows the 3 way nesting structure for MM5 (grid sizes of 0.5°, 0.167° and 0.055°) and 2 way nesting structure of CAMx (grid sizes of 0.167° and 0.055°). The coarse outer domain provided the boundary conditions for the inner domain and inter–relationship was simultaneously simulated. Note that based on the HYSPLIT modeling results the domain of CAMx was extended westwards to cover more upwind regions.

Since the air quality observations are not generally available in Bangladesh, the model was first run on the outer domain (CAMx2) to produce the boundary conditions of air quality for the inner domain (CAMx3). The natural background concentrations of precursors provided by CAMx were assumed as boundary conditions for CAMx2 domain without considerations on temporal variations. Thus, the larger domain (grid size of 0.167°) was simulated with zero initial and boundary conditions. The top boundary conditions used in the model for both domains were obtained from the default values provided by CAMx.

The harmonization between vertical grid structures of both CAMx and MM5 models is provided in Table S5 (see the SM). Vertically, the domain extended from the ground level to 4.177 km and was divided into 5 layers for CAMx and 20 layers for MM5.

Emission input data for CAMx. To prepare Albedo/Haze/Ozone Column (AHO) file to assess the surface reflectance, OMI–TOMS (Ozone Monitoring Instrument – Total Ozone Mapping Spectrophotometer) data available online were collected from NASA (2009).

The emission data for the coarse grid outer domain (CAMx2) were extracted from the data provided by the Center for Global and Regional Environmental Research (CGRER, 2008) of 1° x 1° resolution, available online at (www.cgrer.uiowa.edu/projects/pointer_10_ED.html). The 1° x 1° gridded data were converted to finer gridded (0.167° x 0.167°) data by simply dividing into equal portions without considering other parameters. The emission data for the inner domain (CAMx3) was directly prepared based on the emission inventory results produced in the present study.

VOC profile. There is no VOC specification data available for the study domain or other countries in the region. Therefore, the NMVOC emission profiles developed for the European boundary layer by Kuhn et al. (1998) were used to prepare the relevant profiles used in this modeling study. The lack of local NMVOC profiles is common in Asia hence, for example, a similar approach was also taken by Kim Oanh and Zhang (2002) in a photochemical modeling study for Bangkok, Thailand. The species were prepared in the format of the CBIV chemical mechanism that segregated NMVOC emission into the species groups defaulted in CBIV.

Model performance evaluation. Measurement data, for meteorology and air quality, in the study area were not available to the extent that can be used for a full scale model performance evaluation. Therefore, the model performance was evaluated mainly by analyzing the consistency between the simulated spatial distribution of acid deposition, the meteorological fields (wind and precipitation) and spatial distribution of the precursor emissions. The modeling results were also compared with monitoring values when available.

3. Results and Discussion

3.1. Emission inventory for Dhaka division

Emissions from brick kilns. In Bangladesh, the traditional Fixed Chimney Kiln (FCK) technology is used generally having a low energy efficiency. There were about 3 000 brick kilns in the Dhaka division, of which 60% was coal fired kilns, producing 3 000 millions of bricks in 2006 (BBS, 2008; BMA, 2008). BMA (2008) estimated that about 300 tones of coal and 180 tones of fuel wood were used to produce 1 million bricks.

SO_2 is the dominating pollutant emitted from brick kilns because of the high sulfur content of the coal used (5% by weight). Table 1 shows the results of EI for different pollutants from brick kilns in 2006.

Emission from urban traffic. In the Dhaka division, CNG, gasoline and diesel powered vehicles are running on roads at a high density. CNG vehicles are mostly used in the Dhaka city due to the fuel availability. The ban on 2 stroke baby taxis (Diesel 3W in Figure 3) in Dhaka city and its periphery has been effective since 2003 (DMP, 2006). Hence their number was insignificant in the study area. As seen in Figure 3, CO was the pollutant emitted with the highest

Table 1. *Estimated emissions (t/d) from all sectors in Dhaka division for 2006*

Sector		SO_2	NOx	CO	NMVOC	PM_{10}
Brick kiln[a]	Dry	295	37	94	3.5	35
	Wet	-	-	-	-	-
Transport[a] (annual)		1.3	38	70	27	3
Other sectors[b] (anual)	Non–metallic minerals	9	10	5	0.64	
	Manufacturing industries (Non–specified)	22.5	13	4.7	0.9	0.7
	Railways (trains)	0.4	2.3	0.6	0.3	0.3
	Commercial/Institutional		0.4			
	Residential	19.5	37	1 975	213	57
	Agriculture/Forestry/ Fishing	2.1	2	15.3	3	
	Application of N–containing fertilizers		18			
	Burning of agricultural crop residues	4.3	21	523	39	38
	Waste burning	0.7	4.2	59	4.7	27
Daily average emission (t/d)	Dry	355 (86)[c]	183 (56)[c]	2 746 (51)[c]	293 (53)[c]	161 (51)[c]
	Wet	60 (14)[c]	146 (44)[c]	2 652 (49)[c]	289 (47)[c]	126 (49)[c]
Total annual emission (t/yr)		75 295	59 987	984 994	106 209	52 325

[a] *Calculated based on source and fuel characteristics and their emission factors. Details for vehicle emissions are given in Figure 3.*
[b] *Projected emissions for 2006 based on emission inventory by Hasan (2000) prepared under Male declaration project of UNEP for the year 2000*
[c] *Share in annual emissions (%)*

amounts from the transportation activities followed by NO_x, PM and SO_2. Emissions of CO (and VOC) were mainly from gasoline and CNG powered vehicles while NO_x, PM and SO_2 were mainly from diesel powered vehicles. The large CO amount emitted from the light duty CNG vehicles was mainly caused by its large VKT (almost 1.14×10^9 km/yr) while the large CO emission from the heavy duty CNG vehicles was mainly due to its high EF (24 g/km, Table S3, SM). Diesel powered vehicles generally had lower VKT than CNG vehicles but the high EF of NO_x (8.5 – 17 g/km) resulted in its largest NO_x emission share.

Projected emissions for other sectors. Other sectors included here are industry, agriculture, railway trains, as well as commercial /institutional, residential, open burning activities that were reported by Hasan (2000) for 2000. Industries in Bangladesh use different fuel types such as diesel, furnace oil and natural gas depending on their operations. The residential sector mainly uses LPG, kerosene and biomass for cooking. For the purpose to harmonize the overall EI for the model input over the selected domains we projected the emission from 2000 to 2006 using the sectoral growth (Table S4, SM). Thus, in general all the sectoral emissions increased from 2000 to 2006 except for the pulp and paper industry that reduced by 44% due to its reduced production. The largest increase was estimated for the oil refinery and manufacturing industries (about 60%) and the smallest increase was for agriculture related emissions (15 – 20%). For other sectors, an increase by 30 – 40% was estimated.

Total emissions. Table 1 presents the EI results from this study together with the projected emissions for other sectors in tones per day. Overall, the CO emissions were the highest, above 2 650 t/d in average while emission rates of other pollutants were an order of magnitude lower. Sector–wise, the brick kilns contributed the largest SO_2 emissions (70%), while residential combustion contributed the highest amount of CO, NMVOC (over 72%) and substantial PM_{10} (about 35%), followed by agricultural crop residue burning. NO_x emission was rather uniformly shared by sectors with the highest contribution from the transport and residential cooking, followed by burning of crop residue, application of N–containing fertilizers, and brick kilns.

For the purpose of 3D modeling the hourly emission input is ideally required. In this study, due to the lack of such detail data we first separated the emission for the dry and wet season and further calculated the daily emission as the seasonal average. Many sources listed in Table 1 may be strongly seasonal, e.g. agro–residue open burning or waste burning, but no detailed data were available for a better temporal description. The brick kiln emissions were the only source presented in Table 1 that appeared with a sharp seasonal division as the kilns were not operated during the wet season (because of rain and flooding). Thus, the difference between dry and wet season daily emissions are presented in Table 1 was caused by the brick kiln emission only. Accordingly, SO_2 which was mainly contributed by brick kilns, showed the largest seasonal fluctuation with the emission rate of 355 t/d in the dry season and 60 t/d during the wet season.

Gridded emission distribution for Dhaka division. The gridded emission of SO_x and NO_x for the study area (Figure 4) was prepared based on, as much as possible, the actual distribution of sources on the grids (0.055° x 0.055°). For brick kilns, the location of sources was collected from Bangladesh Bureau of Statistics (BBS, 2008) and the Brick Manufacturers Association of Bangladesh (BMA, 2008). The transportation related emission distribution was prepared according to the traffic volume recorded in different locations of the study area by our primary survey. The survey was conducted in 8 locations (5 for major city inner roads, 2 in residential areas in Dhaka city and 1 for major road outside the Dhaka city) which counted vehicles at each location at morning rush, noon and evening rush hours of a selected day. The survey was done in December 2008 hence it was assumed that the distribution of vehicle density was the same as in 2006. The vehicle densities were used as the weights to distribute the traffic emission over the study domain in producing the gridded emission input file. For other sources (industrial, agricultural, commercial and residential), the land use data taken from Bangladesh Bureau of Statistics (BBS, 2008) and emission from the sources was assigned into the grid based on land use characteristics.

Figures 4a and 4c reveals that in dry season high SO_x and NO_x emissions appeared at the SW corner of the domain, which coincided with the presence of operating brick kilns. During the wet season, both SO_x and NO_x emissions were low over the domain with a higher SO_2 emission rate over the Dhaka city. For NO_x, however, the emission rate was still higher in the western part of the domain (similar to the dry season) which may be attributed to more intensive emissions from agricultural activities in this area. The hourly average calculated from the daily gridded emission of SO_2, NO_x, CO and speciated NMVOC was used for CAMx–MM5 simulation presented in Section 3.2.

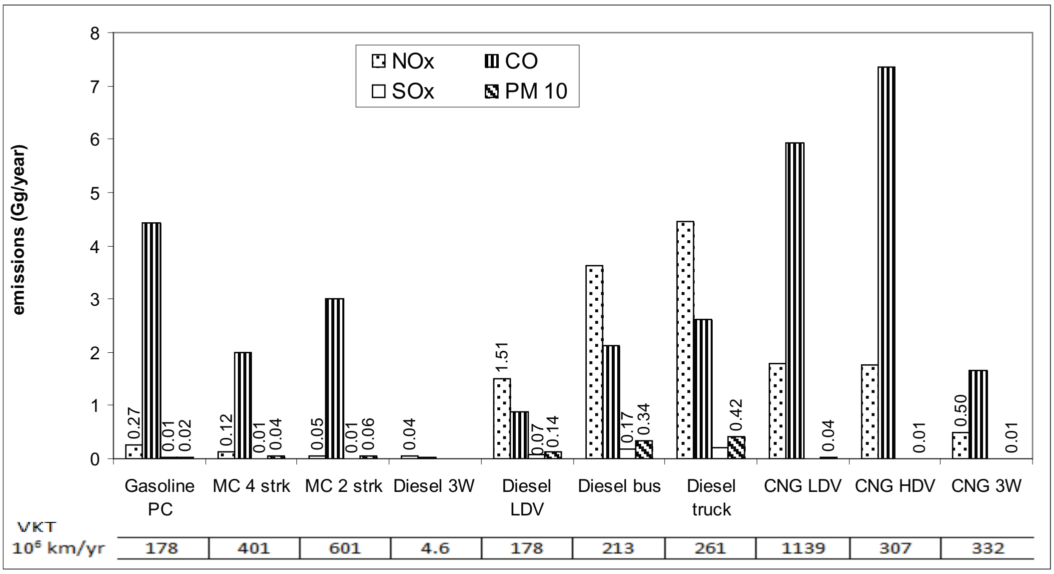

Figure 3. *Vehicle emissions in Dhaka division in 2006. (HDV = Heavy Duty vehicles; LDV = Light Duty vehicles; MC = Motorcycle; PC = Private car; 3W = 3 wheeler).*

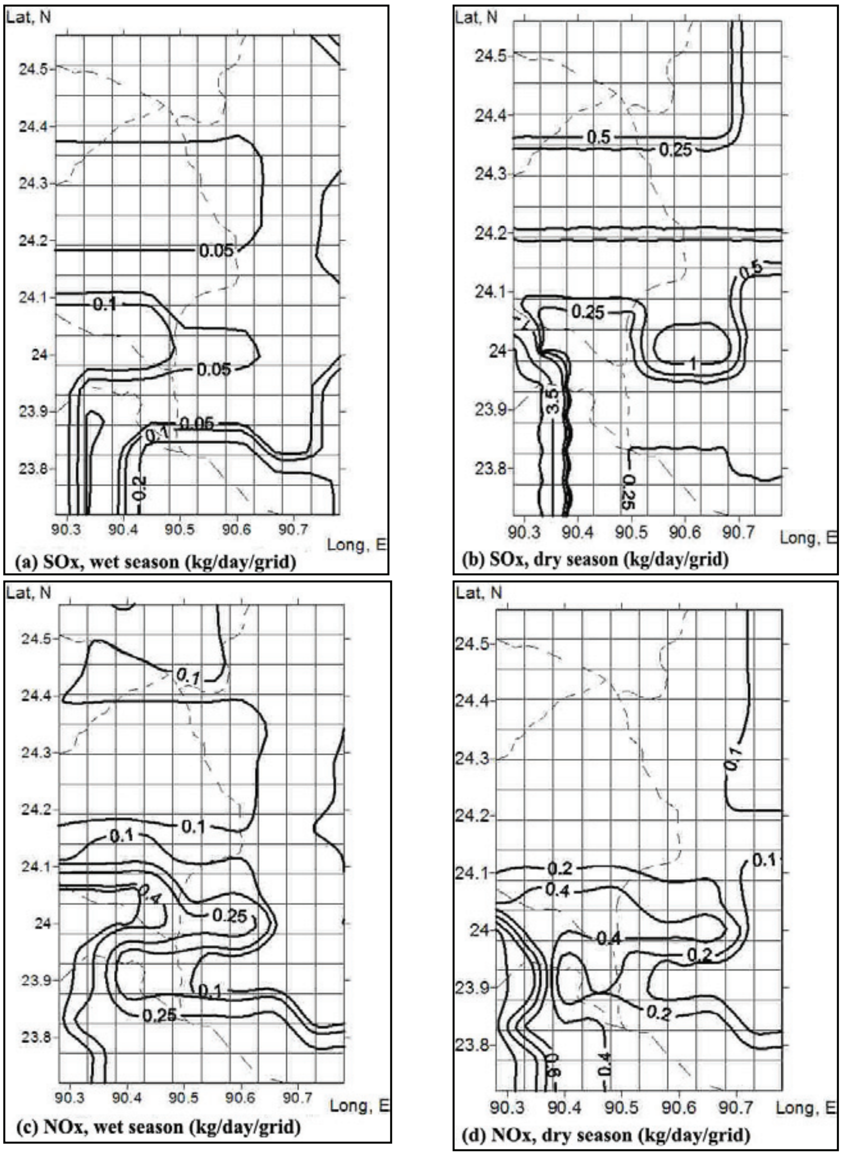

Figure 4. *Gridded emission (kg/day grid) distribution in Dhaka division in 2006, (**a**) SO_x, wet season, (**b**) SO_x, dry season, (**c**) NO_x, wet season, and (**d**) NO_x, dry season.*

3.2. Modeling the acid deposition over Dhaka

Origin of air masses arriving to Dhaka. Figure 5a shows the 6 cluster means of the 10 day back trajectories for the whole year of 2006 produced by HYSPLIT with the dominance of the western origins of air masses arriving at Dhaka throughout the year. For the modeling time period, June and December months, the trajectory patterns are shown in Figures 5b and 5c.

During December, air masses were mainly originated in the continent and had a long pathway through the northern India, before entering Dhaka from north. This continental pathway explains the dry weather in winter. During June, air masses were also originated from the west but over Indian Ocean, they passed India and entered the domain from south. Due to the change in entrance directions of air masses to Dhaka, the local wind shows a distinct difference between the two seasons. During the dry season, the local wind is mainly northerly to northwesterly, while during wet season wind is mainly varied between southerly to southeasterly.

The pathways of air masses reflect the topography of the country with the long Himalaya range situated in the northern part of Bangladesh and hilly regions on the eastern, northeastern and southeastern parts (Figure 1). This topography would channel the air masses to Bangladesh from the west. Thus the large scale movement would be dominated by the west direction all the year round, while the local wind directions are characterized by the seasonal change (windroses inserted in Figures 5b and 5c). To capture the major sources in the upwind locations of the Dhaka division, as mentioned above, the CAMx–MM5 domain was extended westward (Figure 1).

Simulation of acid deposition using CAMx–MM5 modeling system. The simulation results for 2 months (June and December) were used to estimate the respective seasonal deposition rates that were roughly 6 times of respective monthly deposition, assuming similar meteorological conditions and emission over a season. The annual deposition separated in the dry and wet deposition as well as the total (sum of dry and wet deposition) of S and N is presented in Figure 6. In this study, the S deposition included SO_2 and PSO_4 while the N deposition included NO, NO_2, PNO_3 and HNO_3.

The annual average of S and N over the Dhaka domain (summation of the dry and wet season domain–wise) would be 7.6 kg/ha yr of sulfur and 53.7 kg/ha yr of nitrogen. Note that the annual average N deposition (Figure 6f) was quite evenly distributed over the domain with a maximum appeared in the NE part of the domain which reflected the distributed nature of the

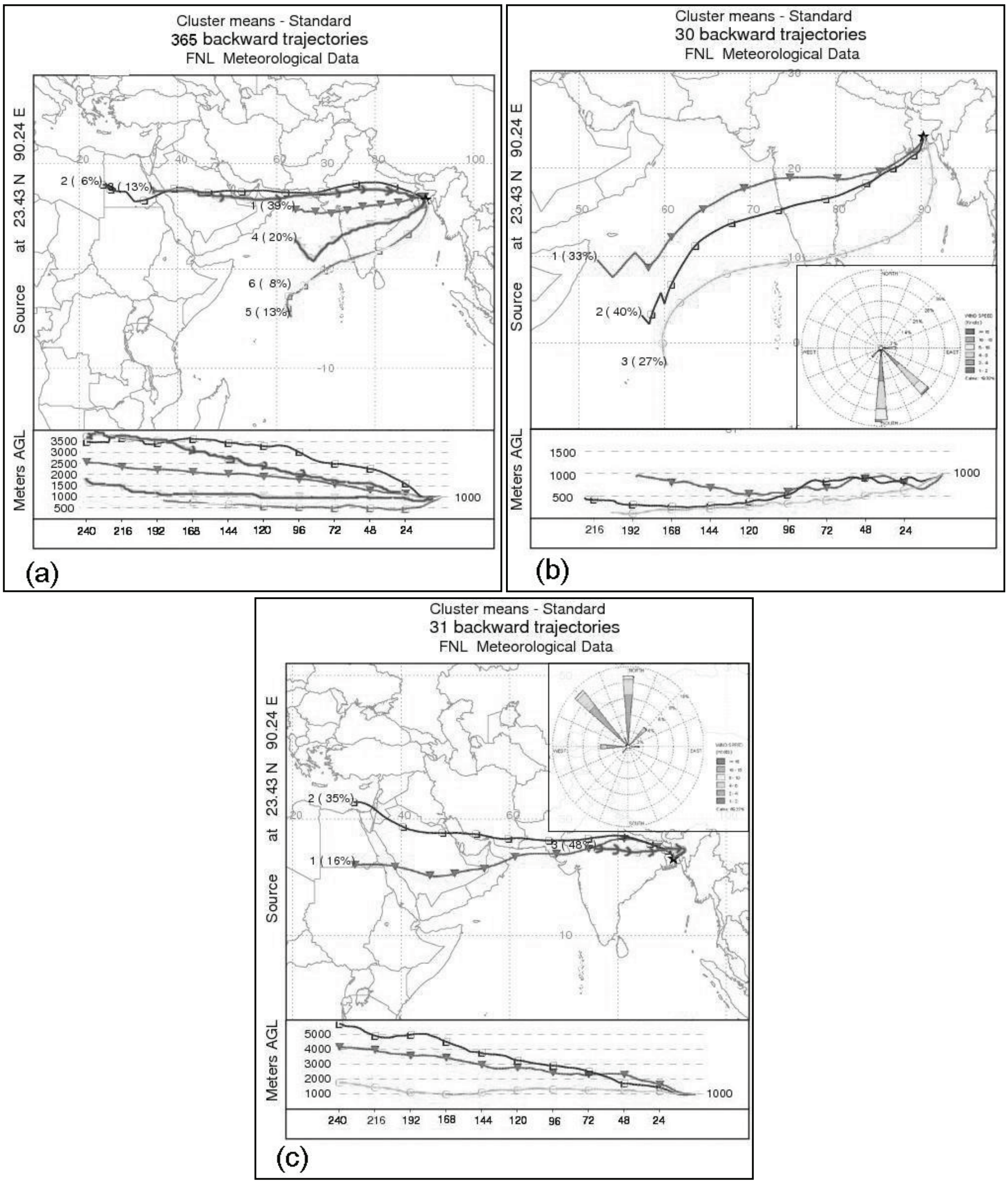

***Figure 5.** Origin of air masses arriving to Dhaka, Bangladesh in 2006 produced by HYSPLIT (10 day backward). (**a**) 6 Cluster means of 365 trajectories of 2006, (**b**) 3 cluster means for the month of June 2006, and (**c**) 3 cluster means for the month December 2006.*

dominant NO_x sources (Table 1). The S deposition (Figure 6c), however, was mainly concentrated in the SW corner where the brick kilns and their high SO_x emissions were located during the dry season (Figure 4b). Overall, the maximum gridded average sulfur deposition rate in 2006 over the domain Dhaka division, 56 kg/ha yr, was lower than that of nitrogen, 77 kg/ha yr. Fortunately these maximum values of S and N deposition appeared in different parts of the domain (Figure 6) hence reducing the peak of total (S+N) deposition on a particular grid (Figure S1, see the SM). Domain–wise the highest annual S+N deposition, in equivalent, was observed in southwest part. Some northeast and central parts of the domain also showed higher annual S+N deposition.

For sulfur, a higher dry deposition was observed as compared to the wet deposition (Figures 6a–6c). The maximum average sulfur dry deposition rate for 2006 in the domain was 56 kg/ha yr, while that of wet deposition was 2 kg/ha yr. The maximum rate of sulfur deposition per season (6 months) in wet season was 6 kg/ha season, while that in dry season was 54 kg/ha season. This suggests a causal link to the dominant contribution to SO_2 emissions by brick kilns that are only observed in the dry season. The same picture was not observed for the simulated nitrogen deposition (Figures 6d–6f). The annual average wet deposition of nitrogen in the domain was higher, maximum of 68 kg/ha yr, than the dry deposition, maximum of 19 kg/ha yr. Deposition of nitrogen was higher in the wet season, maximum of 76/kg/ha season than the dry season, maximum of 4 kg/ha season.

3.3. Model performance evaluation

Acid deposition: The acid deposition monitoring in Bangladesh has not been studied in a large extent so far. Thus, there was no published data to compare with the modeled results. The

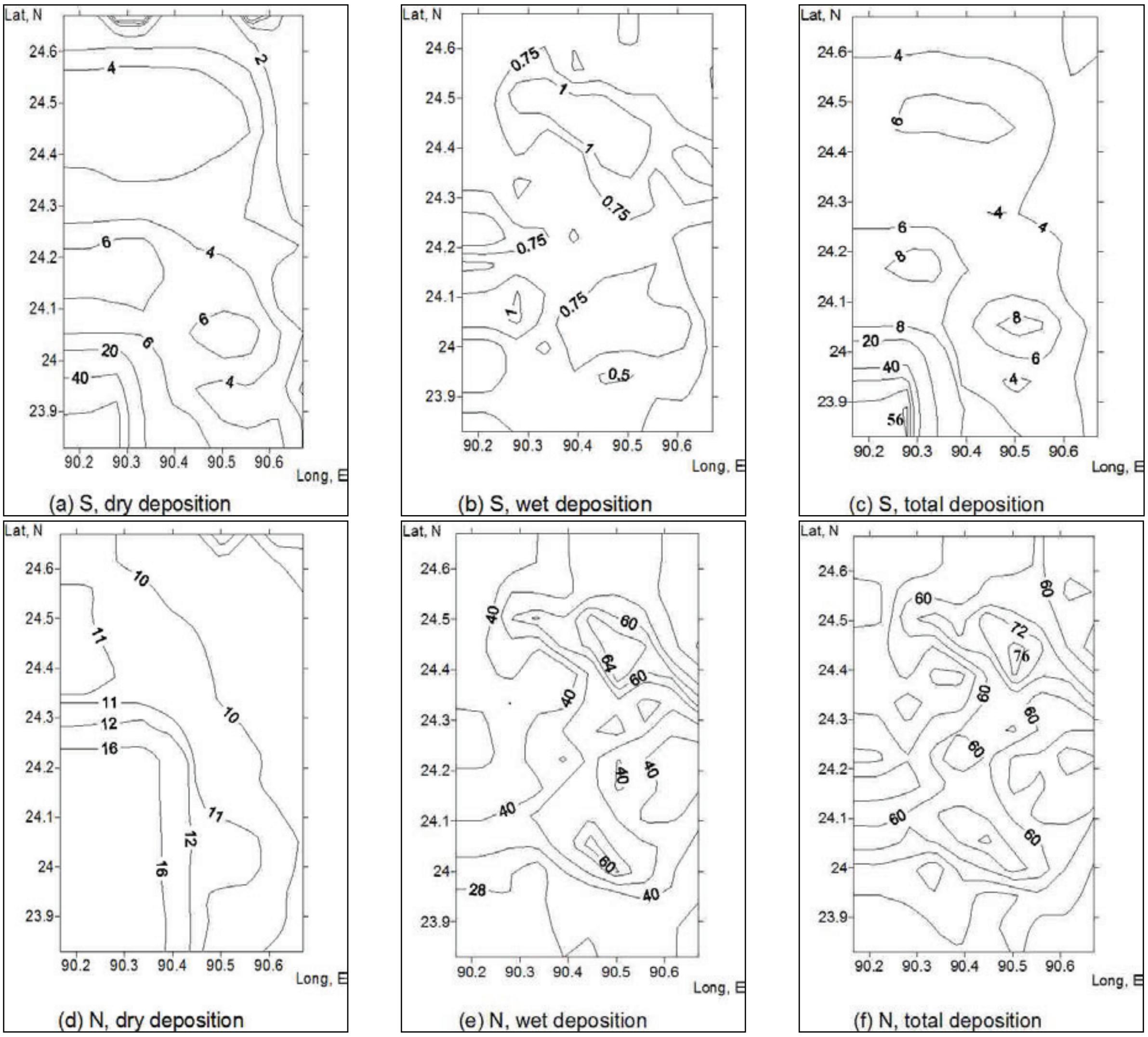

Figure 6. *Simulated distribution of surface S and N annual deposition (kg/ha yr) in Dhaka division for the year 2006.*

projected sulfur deposition (nitrogen deposition was not considered) for Bangladesh for 2005 by RAINS–Asia (IIASA, 1994) was 0.95 kg/ha yr for the country is lower than the simulated average S deposition of 7.6 kg/ha yr by CAMx in 2006 in the present study. Note the IIASA (1994) result was based on the simulation of the projected emissions from all countries in Asia from 1990 to 2005 under the baseline emission scenario (Business As Usual), which may be quite different from the Dhaka emissions in 2006 estimated in the present study. Also, S deposition projected by IIASA (1994) was the country average that would be significantly lower than that over the Dhaka division where emission rates are be higher. Also, the inclusion of the brick kiln emissions in this study significantly increased the domain average deposition. Without the elevation of S deposition at the SW corner (Figure 6c) the domain of S deposition would be still about 4 – 5 kg/ha yr, i.e. about 4 – 5 times higher than the projected data. Thus, the S emissions obtained in this study appeared to be reasonable considering all these points. In any case, monitoring data should be gathered for a meaningful model performance evaluation.

The model results can also be qualitatively evaluated considering the consistency in the spatial distribution of deposition and meteorology fields. The spatial distribution of the simulated acid deposition showed reasonable agreement with the simulated meteorological fields (wind direction and precipitation). In both seasons the simulated deposition was higher in the downwind region (Figure S2, see the SM). In dry season, when the northerly winds prevail, higher deposition was observed in S to SW corners of the domain, which happened to be also the area of the maximum sulfur emissions. In contrast, during the wet season wind direction is mainly from the south hence the deposition was higher in the northern parts of the domain. In fact, the deposition spatial pattern during the wet season showed a clearer dispersion plume as the emission over the domain was rather evenly distributed. During this season, the precipitation rate is a dominant factor for acid deposition. Thus, in the wet season, the spatial distributions of the deposition (Figure 6b and 6e) had some high values in the center of the domain that are also consistent with the precipitation pattern (Figure S2, see the SM).

SO_x and NO_x concentrations. The simulated spatial distribution of monthly average concentrations of SO_2 and NO_x, in December and June is given in Figure 7. Overall, the SO_2 concentration was higher where higher emissions, i.e. in the places where industries, brick kilns and diesel vehicles were prominent. In December, the simulated SO_2 and NO_x concentrations reached above 70 ppb and 100 ppb, respectively, at the SW corner of the domain where brick kiln emissions were high. In June the maximum SO_2 and NO_x concentrations, above 15 ppb and 70 ppb, were observed over the Dhaka city (south of the domain).

The CAMx model results were compared with the annual average monitoring results of SO_2 and NO_x available in Dhaka for

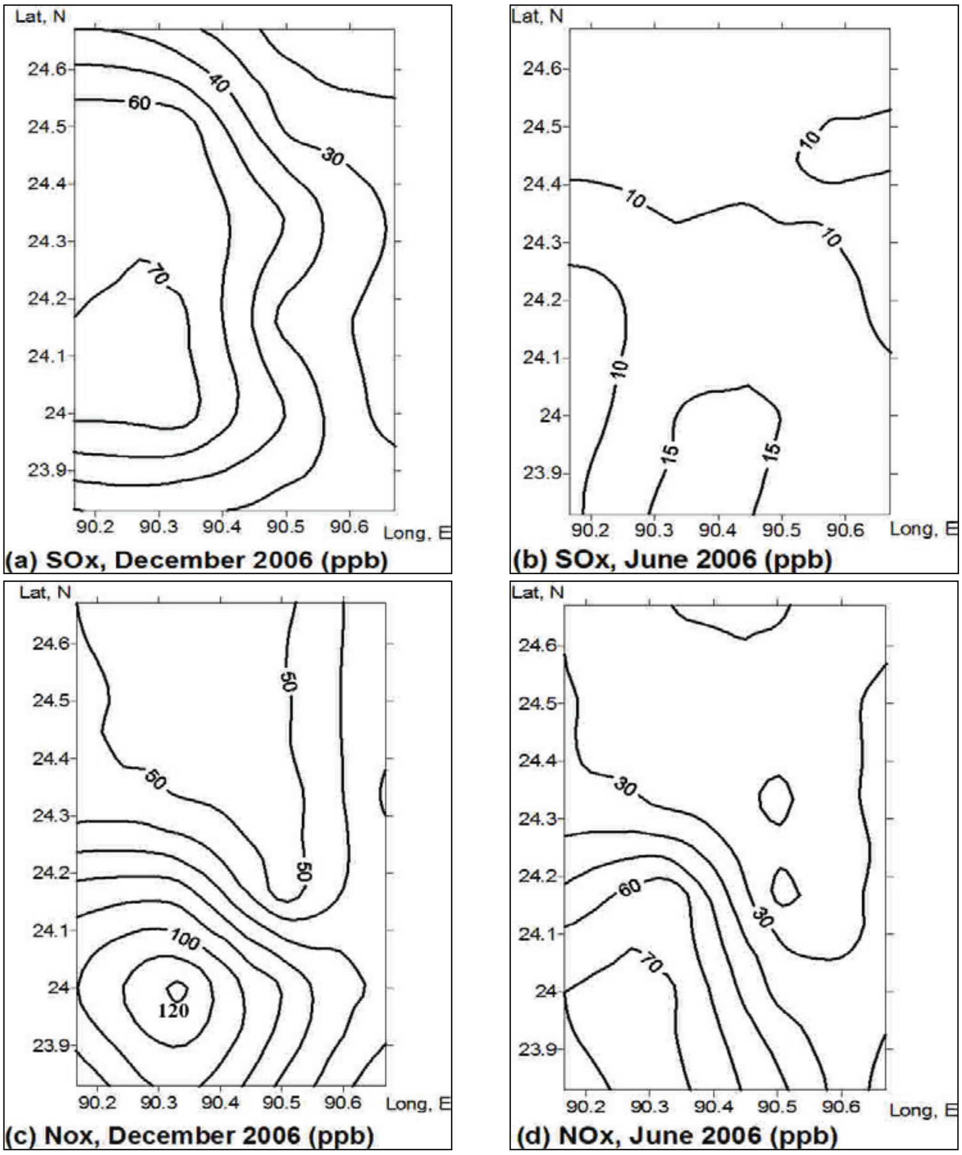

Figure 7. *Distribution of monthly average surface SO_2 and NO_x concentrations (ppb) over Dhaka division by CAMx model for 2006.*

2004 and at a background site (rural area outside of the domain CAMx3) for 2006 (Table 2). There was no continuous monitoring data reported for the Dhaka division and these appeared to be the only accessible monitoring data to evaluate the model results. The simulated annual concentrations of NO_x in the Dhaka city were in the same range of the monitoring annual average of 2004 which was estimated based on measurements at 6 sites in the city (SOE, 2005). However, the simulation results were somewhat higher than monitoring data for SO_2. In fact with the large sectoral growth discussed above the simulated results for an increased emission in 2006 are expected to be higher than the monitoring results in 2004. However, many factors may cause the differences between monitoring and modeling. These are related to the uncertainty in the emission estimates, the gridded model outputs and the scarcely distributed monitoring points. The simulated as well as monitored results for Dhaka city were well above the monitored levels in a clean background site of Bangladesh in 2006 (Table 2).

Meteorology. The MM5 model output was compared to the monitoring data of Bangladesh Meteorological Department, at the Dhaka station (Lat 23°9', Long 90°24') for June and December 2006 and showed a reasonable agreement in terms of average wind speed (1.17 m/s by modeled vs. 1.23 m/s by monitored in December; 1.94 m/s by modeled vs. 2.1 m/s by monitored in June), average daily maximum temperature (23.3 °C vs. 26.8 °C in December; 29.6 °C vs. 32.3 °C in June) and average daily minimum temperature (16.7 °C vs. 15.8 °C in December; 22.8 °C vs. 26.1 °C in June) (Table 3). Extensive statistical analysis would be desirable when sufficient monitoring data are available.

Table 2. *Comparison between annual average monitored and modeled concentrations (ppb) of SO_2 and NO_x in Dhaka city, Bangladesh*

Pollutants	Simulated, Dhaka city (2006)	Monitored		Bangladesh Annual standard [a]
		Dhaka city, 2004[a]	Background site, 2006[b]	
SO_2	27±21	12±8	1.54	30
NOx	72±26	70±58	2.15	53

[a] *SOE (2005),* [b] *Male (2009): rural area of Satkhira, outside the Dhaka Division*

Table 3. *Comparison of simulated meteorological output of MM5 with monitoring station data of Bangladesh Meteorological Department for 2006 (monthly average)*

Parameters	**MM5**	**BMD**
Monthly avg. Max Temp (°C)		
December 2006	23.3	26.8
June 2006	29.6	32.3
Monthly avg. Min Temp (°C)		
December 2006	16.7	15.8
June 2006	22.8	26.1
Monthly avg. Wind Speed (m/s)		
December 2006	1.17	1.23
June 2006	1.94	2.1
Suggested criteria (%) [a]		

Local and regional contribution to acid deposition in Dhaka division. As described above, the outer domain (CAMx2) covers the most of Bangladesh and some adjacent areas (Figure 1) for which the emission input was drawn from CGRER. Considering the zero emissions from the Dhaka division in both dry and wet season, the CAMx simulation shows that the deposition resulted from the emissions from domain CAMx2, outside Dhaka division, contributed approximately 10% and 19% of total deposition over Dhaka in the division in dry and wet season, respectively. Thus, the local emissions, from the Dhaka division itself, appears to be mostly responsible for the acid deposition in the study area. The spatial distribution of the acid deposition in Dhaka originated from emissions outside the domain is shown in Figure S3 (see the SM).

Note that, the CAMx2 domain mainly covered the Bangladesh territory, the contribution from outside Dhaka (outside CAMx3 domain) in this case was still the domestic emissions (from other parts of the country) to the Dhaka division and does not reflect the contributions from upwind regions outside the country. Thus, the lower emissions from other parts of the country, as compared to Dhaka division, have resulted in their lower contributions to acid deposition over the Dhaka division. A larger domain should be selected to study the long–range transport from upwind regions outside the country to the study area.

3.4. Discussion: Toward assessment of acid deposition impact in Bangladesh

SRDI (2008) reports that the observed pH of soil in Dhaka division ranges from neutral to slightly acidic, which implies that, a reasonably high acid deposition in the area could cause major impacts on the ecosystem. The critical load defined by IIASA (1994) for the ecosystem of the country is 1 500 eq/ha yr.

The preliminary deposition results in this study show that in Dhaka division the highest deposition in the domain can reach 6 450 eq/ha yr while the domain average value was 3 640 eq/ha yr, i.e. well exceeded the critical load. The main deposition, averaged over the domain, was 3 240 eq/ha yr for nitrogen while sulfur deposition was only 400 eq/ha yr. The main deposition occurs during the wet season since the total N+S deposition rate was 390 eq/ha season, during the dry season. Sulfur dry deposition was higher during the dry season with the peak values in the SW corner of the domain due to the emissions from brick kilns as mentioned above. N deposition is, however, significant with the highest deposition in the wet season. Even though N is an important nutrient in soil for ecosystems, too high N deposition would cause harmful effects.

Our results presented earlier show that the simulated acid deposition reasonably reflects the seasonal changes in the precursor emissions and meteorology. However, the ratio of N to S deposition (N/S) was not proportional to the NO_x and SO_x emissions. In the dry season, the daily SO_x emission over the domain was about 2 times higher than NO_x (Table 1). However, the N and S depositions were almost in the same range. During the wet season, the emission for NO_x was only about 2.5 times higher than SO_x but N deposition was significantly higher than S deposition (50 – 60 times). The high wet deposition of N may be also linked to a more efficient wet removal process associated with high precipitation rates. In addition, the chemistry of acid rain precursors (SO_x and NO_x) incorporated in the chemical mechanisms used in the model is also important to explain the difference in the deposition rates of sulfur and nitrogen. Wang et al. (2008) studied the acid deposition simulation using eight different chemical transport models and showed that different models produced different results. Some models overestimated while others underestimated the deposition, which depends on the reaction rate coefficients of precursors. Due to lack of the acid deposition monitoring data to compare with the simulation results, the modeled values in this study are considered tentative. Further studies should focus on gathering the monitoring data, refine the emission inventory and also simulation with other models to improve the modeling results.

Nevertheless, the risk of high acid deposition may be real, especially with the current increasing emission sources. Hicks et al. (2008) assessed the acid deposition scenarios over South and Southeast Asia along with the potential of the region to reach the 20% base saturation level. The study showed that with the rapid economic growth and with no further emission control, more acid deposition and more soil acidification is expected in the country, and in 25 years some parts of Bangladesh would reach the 20% base saturation level. Therefore measureable impacts would be induced on ecosystems. However, extensive investigation and detailed critical load assessment for the local ecosystem is required before making a concrete statement on the impacts.

4. Conclusions

The HYSPLIT back trajectories showed that most of air masses arriving to Bangladesh were originated from the west in a year round. The directions of air masses to Dhaka, however, changed abruptly with seasons bringing the local wind directions from NW–N in the dry to S–SE in the wet season. In the dry season the long continental pathway may bring in considerable LRT air pollution to the country.

Brick kilns, operated only during the dry season, contributed significantly to SO_2 emissions of Dhaka division, 295 t/d. Annual average daily emissions, from the division for SO_2, NO_x, CO, PM_{10} and NMVOC were 207, 165, 2 699, 283 and 152 t/d, respectively. The simulated annual average of S and N over the domain (summation of the dry and wet season domain–wise) was 7.6 kg/ha yr of sulfur and 53.7 kg/ha yr of nitrogen. Overall, the simulated acid deposition was reasonably related to local emission distribution and precipitation rate. Without consideration of the emissions from the neighboring countries, more than 80 – 90% of the acid deposition in the Dhaka division was from the local emissions while the rest was associated with the domestic emissions from other parts of the country. The preliminary simulation results show that the high total (N+S) annual acid deposition rate in 2006 may already present a threat to the local ecosystem. The high economical growth of the country, coupled with a low degree of emission control, would lead to the increase in the future emission hence more threat would be expected.

In order to provide a more realistic assessment of the acid deposition threat in the country further studies are required to update the EI with detailed activity data and revised emission factors for anthropogenic sources. Natural sources should also be included. The model domain should be extended to assess the contribution from other upwind countries. Monitoring of rainwater acidity and its comparison are required for modeling performance evaluation.

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Supporting Material Available

Emission factors used in EI for brick kilns of Bangladesh in 2006 (Table S1), Vehicle population and VKT of Bangladesh and Dhaka division in 2006 (Table S2), Vehicle EF used for the EI of transportation sector (Table S3), The country sectoral growth factors used in upgrading emission inventory from 2000 database to 2006 (Table S4), Harmonization of vertical structure between MM5 and CAMx (Table S5), Total N+S deposition in eq/ha per dry season (a), wet season (b) and annual (c) (Figure S1), Simulated meteorological conditions (wind direction with precipitation), dry (represented by December simulation results) and wet (represented by June simulation results) season acid deposition in Dhaka division of Bangladesh (Figure S2), Simulated total acid (S+N) deposition in Dhaka division of Bangladesh with only emission outside Dhaka division (a) dry season, 2006 (b) wet season, 2006 (Figure S3). This information is available free of charge via the Internet at http://www.atmospolres.com.

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