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STUDY ON THE SULFUR DIOXIDE POLLUTION IN THE CAPITAL CITY OF A DEVELOPING COUNTRY IN WINTER

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Abstract

Ten-day-average concentrations of SO_2 were measured using molecular diffusion tubes in Dhaka, Bangladesh in winter 1995-96, and its spatial distribution and temporal variation were simulated using an Eulerian transport/chemistry/deposition model.
The measurement first elucidated the characteristics of the spatial distribution of SO_2 in this area, and showed extremely high concentrations in the south-eastern industrial zone of Dhaka where the highest concentration was about 100 ppb. Motor vehicles and brick fields were speculated two major emission sources. The model well reproduced the observed concentrations. The chemical conversion rate of SO_2 to SO_4^{2-} over Dhaka was estimated, and it showed large diurnal and spatial variations. The average pseudo-first-order-reaction coefficient of SO_2 to SO_4^{2-} at ground level in winter was about 0.3 % h^{-1}, resulting in 7% conversion in a 24-hour period. The spatially averaged dry deposition of SO_2 was 112.5 mol km^{-2} in 10 days over Dhaka.

KEYWORDS: *Developing country, Dhaka, SO_2, measurement, simulation*

1. Introduction

Atmospheric pollution in urban area is a major issue in many developing countries all over the world. Dhaka, the capital of Bangladesh having a population of about 9 million, is one of the biggest cities of the developing countries. The city is also a treasury of inestimable monuments of the history and culture of Bangladesh. But in recent years, Dhaka is experiencing severe air pollution (especially SO_2) in winter (Azad and Kitada, 1996). The highly elevated pollutant concentrations have already threaten public health, vegetation, and invaluable ancient monuments.

Emissions from various peculiar kinds of diesel traffic vehicles and badly maintained automobiles contribute most to air pollution problems. Brick fields, which are grown up abundantly around Dhaka to meet the increasing demand for construction materials, and use coal as main fuel, are another major contributors to the severe air pollution in Dhaka.

To develop a reliable control strategy it is necessary to know the present pollution levels so that diffusion tube samplers were used for monitoring SO_2 concentrations in Dhaka. The

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samplers require no maintenance or power supply since they collect SO_2 via molecular diffusion. This method has been extensively used for measurements of ambient SO_2 and NO_2 concentrations in both urban and rural areas (Atkins *et al.*, 1986 ; Bower *et al.*, 1991 ; Gair *et al.*, 1991 ; Maeda *et al.*, 1994).

The development of efficient control strategies for air pollution problem in urban area should also be based on a better understanding of the physical and chemical processes that govern the formation, transport, diffusion, chemical transformation and removal of the pollutants. Thus, mathematical model, since it provides the necessary analytic framework for describing the physics and chemistry of polluted urban airsheds and simulating urban air pollution episodes, has been extensively used to calculate the distribution of air pollutants and to formulate effective strategies for controlling the air pollution (Juda, 1986 ; Nester and Fiedler, 1992 ; Pilinis *et al.*, 1993 ; Kumar and Russell, 1996).

This paper presents the results of measurements of SO_2 concentrations in Dhaka, and also reports the application of an Eulerian transport/chemistry/deposition model (Kitada *et al.*, 1984 ; Carmichael *et al.*, 1986 ; Kitada *et al.*, 1993) to simulate SO_2 concentrations over greater Dhaka. An investigation on the chemical conversion of SO_2 to SO_4^{2-} is also performed. These are the first results of measurements and modeling effort in Dhaka. In the context of urbanization, this study will be useful for emission control strategies, decision making processes, planning and management of large cities of the developing countries like Dhaka.

2. Measurements

2.1 The Method

Molecular diffusion tubes were used to measure the concentration distributions of SO_2 at 64 sites in Dhaka city and its suburbs during the period of December, 1995 to January, 1996. In these measurements we adopted the procedures described in Maeda *et al.* (1994), in which sodium carbonate, coated on filter paper, was used to absorb the SO_2 gas from the air. The amount of sulfate absorbed was then determined using Dionex ion-chromatograph (model DX-300), and the results were converted to air concentrations of SO_2 using the exposure period and calibration equation.

The diffusion tube samplers were calibrated using 6 automated air pollution monitoring stations in Aichi-prefecture, Japan. Three samplers were deployed at each automated air pollution monitoring station for 10 days to measure the concentration of SO_2. The average sulfate extracting rate of the three samplers at each station was regressed against SO_2 concentration as shown in Figure 1. Maeda *et al.* (1994) had relatively better calibration line. Slight scattering of our data as could be seen in Figure 1 may be due to the possible effect of wind speed. All three samplers at each monitoring station gave consistent result. It should be noted here that we were forced to extrapolate the regression line when applied to Dhaka data in which the highest SO_2 concentration was as large as 100 ppb.

2.2 Description of Sampling

The sampling was carried out from mid-December of 1995 to mid-January of 1996 at 64 sites in the highly populated city of Dhaka and its suburbs, including Savar, Tongi, Kachpur, Futulla and Keraniganj (Figure 2(b)). Dhaka is at latitude 23°43′ N and longitude 90°24′ E, and in the middle of Bangladesh (latitude 20°34′ to 26°38′ N, longitude 88°01′ to 92°41′ E), a

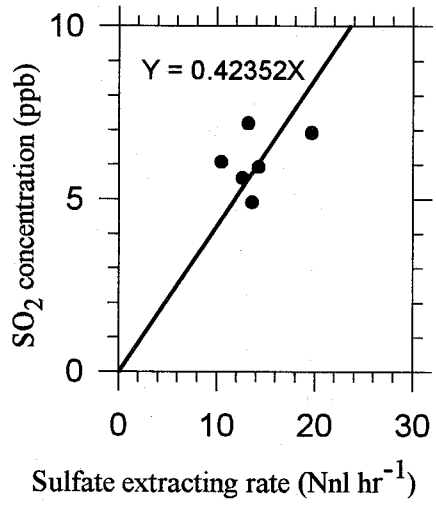


Figure 1. Calibration line between sulfate extracting rate and gaseous SO_2 concentration.

country which lies in the eastern part of south Asia and is bounded by India on the west, the north and the north-east, and Burma on the south-east, and the Bay of Bengal on the south (Figure 2(a)). Dhaka, located in flat plain with no mountain, is surrounded by rivers at all sides and is the centre of commerce in Bangladesh. The city is growing rapidly, but not in a well planned way. The weather and climate patterns are monsoonal.

The sampling sites were selected to reflect ambient concentrations of SO_2 over all types of area and environment in Dhaka. One set of samplers was exposed within 15 $\sim$ 50 m from major roads, another set was set up far away ($>$ 100 m) from major roads. The third set of samplers (largest set) was deployed in the planned and unplanned housing area, commercial area, industrial area, suburban and other types of area to measure ambient concentrations of the pollutant. The locations of the sites are shown in Figure 2(b). The samplers were attached to walls, windows, fences, poles and leafless trees at man height. Three samplers were exposed at a site for 7 $\sim$ 10 days and the samplers were sealed into plastic bag after recapping at the end of the exposure period. Data from three samplers were averaged to give concentration at a site. Almost no sampler showed any unusual behavior.

2.3 Meteorology at the Study Area

Meteorological data were obtained at the Dhaka national observatory (see Figure 2(b)). During the measurement period, the afternoon temperatures were moderately warm ($\sim$ 26°C) and relatively cooler temperatures were observed during the night ($\sim$ 13°C). The maximum temperature was 28°C on the 25th and 31st December, 1995, and the minimum temperature was 9.6°C on the 14th January, 1996. The average temperature was 20°C. Very low wind speeds averaging 0.52 m s^{-1} with 68% calm (wind speed less than 0.5 m s^{-1}) were observed during the study period. The north-westerly was the predominant wind, which blows from the Himalayas to the Bay of Bengal and is typical in this season. The diurnal patterns of wind speed and wind direction in Dhaka for typical two days, for example 26 and 27 December 1995, are shown in Figure 3. There was no rain in the measurement period. The relative humidity approached above 90% on most days just before sunrise. Fog was also observed during most

(a)

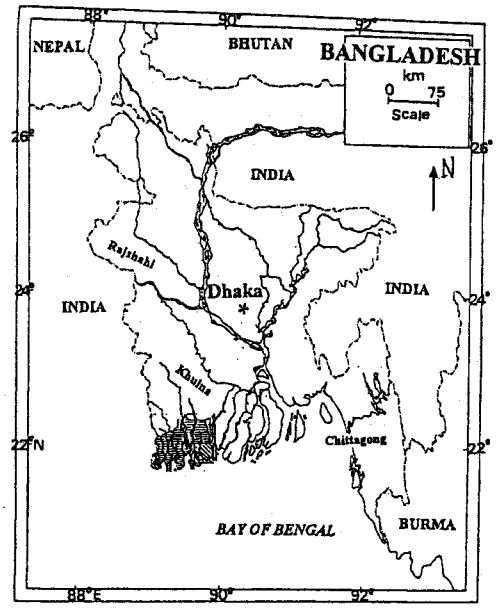
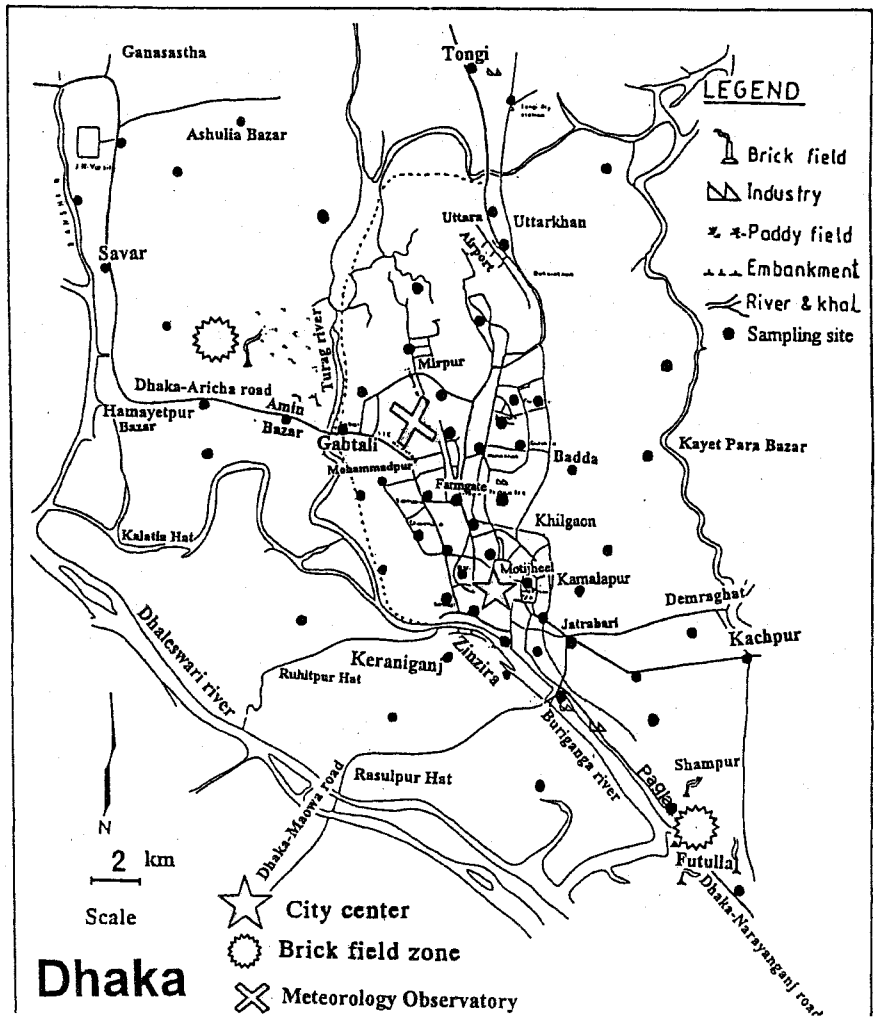


Figure 2. (continue)

(b)



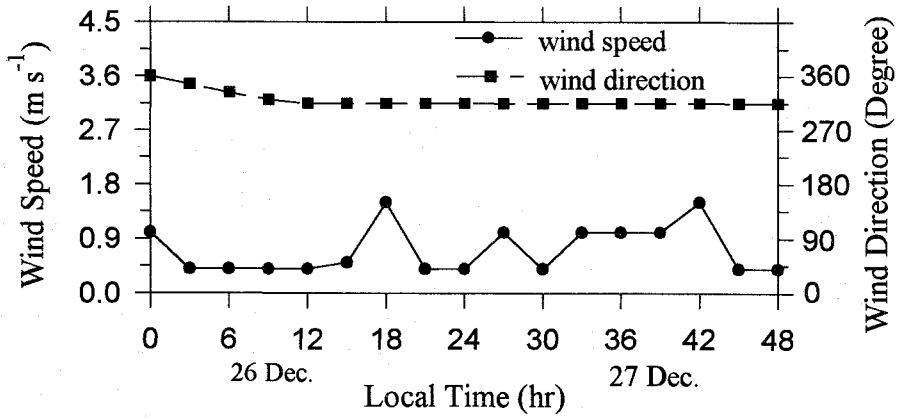


Figure 3. Diurnal patterns of wind speed and wind direction at Dhaka in winter 1995 for two typical days *i.e.*, 26 and 27 December (we assumed 0.45 m s^{-1} wind velocity in calm condition and wind direction was interpolated).

mornings and it disappeared at about 0800 BST (Bangladesh Standard Time).

Mixing height, which controls the vertical dispersion of pollutants, was estimated for winter season using the vertical temperature profiles (Holzworth,1967) from morning (0000GMT) aerological data and the surface temperature at 0600 GMT (1200 Bangladesh Standard Time). We have used the aerological data obtained for January and December, 1989, since those data were available only for that year. The average mixing depth at 0600 GMT in Dhaka in December and January was 700 and 750 m respectively, which were similar to those in New Delhi, India, where the depth was less than 1 km in winter (Gamo *et al.*, 1994).

2.4 Measurement Results and Discussion

(1) Analysis of the Concentration Distributions of SO_2 over Dhaka

Spatial distributions of SO_2 concentrations over Dhaka (Figure 4) show extremely high SO_2 concentrations in the south-eastern industrial and brick field zone, where the highest 10-day-average concentration was about 100 ppb, which is about 13 times larger than that at the polluted site in Nagoya, Japan in the same season. Polluted zone, in which the average SO_2 concentration was over 40 ppb, extended along major road running from north-west to south-east, and also parallel to the Buriganga river in Dhaka area. Brick fields and industries along the traffic and navigation routes as major emission sources, and pollutants-transporting north-westerly blowing along the river as prevailing wind in winter, have formed this particular high SO_2 zone. The 10-day-average ambient SO_2 concentrations and their standard deviations were 18±9 ppb in the city center, 31±24 ppb within the distance of 15 $\sim$ 50 m from major routes and 10±8 ppb in the suburban area. Use of high sulfur containing fuel in traffic vehicles is the main reason of high SO_2 along the major routes. A statistical analysis of the measured SO_2 concentrations at 64 sites in Dhaka is shown in Table 1.

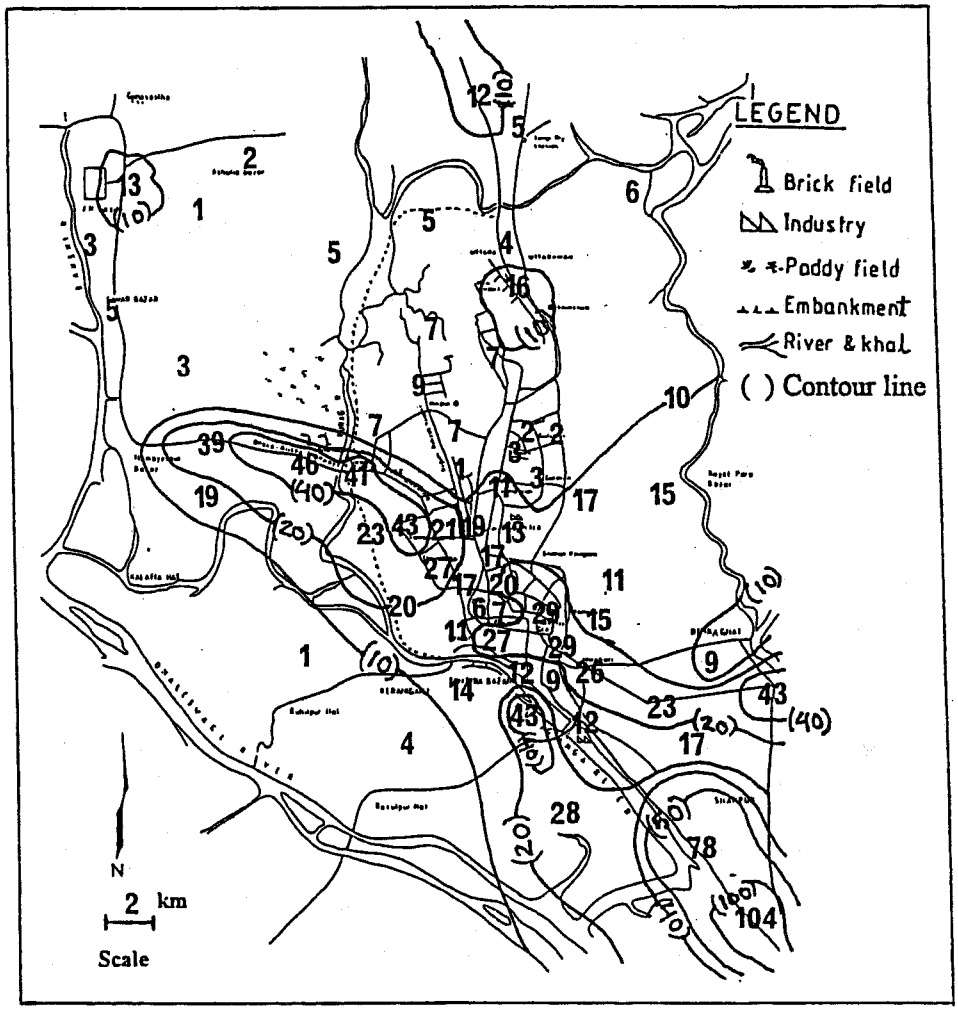


Figure 4. Distributions of ambient SO_2 concentrations over Dhaka in winter 1995 - 96 (the number represent SO_2 concentrations in ppb).

Table 1. Summary statistics of SO_2 concentrations (ppb) in Dhaka in winter 1995 - 96

Pollutant	No. of Site	Mean	S.D.	Range		Percentile						
				Min.	Max.	20	40	60	80	90	95	99
SO_2	64	17.2	17.8	1	104	5	10.4	16	27	41.7	45.7	102.8

Table 2. Sites with 10-day-average SO_2 concentration exceeding 40 ppb

Site	SO_2 Concentration (ppb)
Futulla	104
Pagla	78
Amin Bazar	46
Kachpur	43
Zinzira	43
Mohammadpur	43
Gabtali	41

Table 3. SO_2 concentration levels in some urban areas

City	SO_2 Concentration (ppb)		References
	Maximum	Mean	
Sapporo (Japan)	21	7.4	Maeda *et al.* (1994)
Nagoya (Japan)*	7	6	DOE**, Aichi-Prefecture
Agra (India)	8.4	5.6	Goyal *et al.* (1994)
Varanasi (India)	29.3	20.5	Pandey *et al.* (1992)
Dhaka	104	17.2	Present study

* Based on 10-day-average concentrations at 4 sites in November-December 1995.

** Department of Environment.

(2) Comparison with Air Quality Standards, and with that in other Cities

According to the ambient air quality standards set by the Japanese government (for example, JEA, 1985), daily average of hourly values shall not exceed 40 ppb for SO_2. The limit value for SO_2 (40 ppb) was exceeded at 7 sites (and possibly at an eighth site with a 10-day-average of 39 ppb), where 10-day-average concentration was above 40 ppb (Table 2). These sites are located especially in the vicinity of the south-east, and north-west brick field and industrial areas. The World Health Organization (WHO) guidelines for SO_2, which are 500 μg m^{-3} for 10 minutes and 350 μg m^{-3} for 1 hour exposure, become about 188 μg m^{-3} (*i.e.*, about 66 ppb) after logarithmic interpolation for 10-day-average. This value (66 ppb) was exceeded at 2 sites in the south-east industrial and brick field zone (see Figure 4). It is convenient to present results from studies in other urban areas, in order to give proper significance to the concentrations measured in Dhaka. Table 3, which contains SO_2 concentration levels in some urban areas, indicates that SO_2 in Dhaka city (present study) is relatively very high compared with those in Agra and Nagoya. The maximum SO_2 concentration in Dhaka is more than 2 times higher than that of Varanasi, India. Furthermore, the highest SO_2 in Dhaka is also four times larger than the winter- time maximum concentration of Sapporo, Japan. The measurement procedures and sampling device used here were same as those in Sapporo.

3. Modeling Study

3.1 Model Description

A three-dimensional Eulerian transport/chemistry/deposition model, described in the work of Kitada et al. (1984), Carmichael *et al.* (1986) and Kitada *et al.* (1993) to study urban air pollution, was used to simulate SO_2 concentrations over Dhaka. The equation represents a mass balance in which all of the relevant emissions, transport, diffusion, chemical reactions and removal processes are expressed in mathematical terms as follows :

$$\rho\frac{\partial C_i}{\partial t} + \rho u\frac{\partial C_i}{\partial x} + \rho v\frac{\partial C_i}{\partial y} + \rho w\frac{\partial C_i}{\partial z} = \frac{\partial}{\partial x}\left(\rho K_H\frac{\partial C_i}{\partial x}\right) + \frac{\partial}{\partial y}\left(\rho K_H\frac{\partial C_i}{\partial y}\right) + \frac{\partial}{\partial z}\left(\rho K_v\frac{\partial C_i}{\partial z}\right) + R_i + S_i \quad (1)$$

where C_i represents the concentration of the i th chemical species (dimensionless),
ρ the density of air,
u, v, and w the mean wind velocity components,
K_H and K_v the horizontal and vertical eddy diffusivity,
R_i the chemical reaction term, and
S_i the emission rate of point source of pollutant i

(1) Numerical Solution of Diffusion Equation

The partial differential equation (1) is solved in four steps using the concept of the fractional time steps : (i) advection/diffusion equations are solved in the x-direction, (ii) the same procedure adopted in the y-direction, and (iii) also for vertical advection/diffusion, and then (iv) chemical transformations are calculated for reactive pollutants. The model performs this four-step procedure during each time step. One dimensional finite element method is used for numerical integration in the above (i), (ii) and (iii). An upwinding parameter and spatial filtering are included in the analysis.

(2) Treatment of Advection and Diffusion

Flow fields were estimated using the wind measured at every three hours. Horizontal eddy diffusivity, K_H, was given by the following equation after Pielke (1974),

$$K_H = \alpha^2\,\Delta x\,\Delta y\,[\left(\frac{\partial v}{\partial x} + \frac{\partial u}{\partial y}\right)^2 + \frac{1}{2}\{\left(\frac{\partial u}{\partial x}\right)^2 + \left(\frac{\partial v}{\partial y}\right)^2\}]^{1/2} \quad (2)$$

A lower limit of K_H=1000 $m^2\ s^{-1}$ was imposed.

The vertical diffusivity (K_v) was calculated using the following relation (Tanaka, 1991), which is based on the output of $k-\epsilon$ turbulence model for inland boundary layer (Kitada, 1987) :

$$K_v = \left(\frac{10.417}{h^2} - \frac{0.374}{h}\right)z^2 + \left(0.36 - \frac{5.625}{h}\right)z + 0.013z + 0.208 \quad (3)$$

where h is the mixing height and z is the height above the ground, $z \leq h$; for $z > h$, $K_v = 1$ $m^2\ s^{-1}$ was used. The upper limit of K_v was restricted to 50 $m^2\ s^{-1}$.

(3) Treatment of Atmospheric Chemistry

The spatial and temporal distribution of SO_2 in the atmosphere should be dependent on chemical environment, since important SO_2-oxidizers such as OH radical are formed from NO_x, hydrocarbon and O_3 chemistry. The chemical mechanism used in this model was adapted from the work of Lurmann *et al.* (1986). The mechanism takes 148 reactions among 70 species into account *i.e.*, 44 advected and 26 steady-state species. The mechanism includes major photochemical smog reactions as well as production processes of acidic pollutants.

(4) Treatment of Dry Deposition

Many types of pollutants, including sulfur compounds, nitrogen oxides, and ozone are removed from the surface layer by such features as vegetation through the process of dry deposition. The dry deposition of different species may be described by their dry deposition velocity. It involves a complex linkage between turbulent diffusion in the surface boundary layer, molecular scale motion at the air-ground interface, and chemical and biological interactions of the material with surface. The flux of material i to the ground F_i is defined by:

$$F_i = V_{g,i} C_i \tag{4}$$

where $V_{g,i}$ is the deposition velocity of species, i. In order to parameterize the dry deposition velocity of gases to surfaces the multiple resistance-based scheme is used.

$$V_g = \frac{1}{r_a + r_s + r_c} \tag{5}$$

The aerodynamic (r_a), surface (r_s), and residual (r_c) resistances are defined as :

$$r_a = \{ln\left(\frac{z_r}{z_0}\right) - \psi_c\left(\frac{z_r}{L}\right)\} / (ku_*) \tag{6}$$

$$r_s = ln\left(\frac{z_0}{z_d}\right) / (ku_*) \tag{7}$$

$$r_c = \frac{1}{k_c} \tag{8}$$

where z_r represents the reference height where V_g is defined,

z_0 roughness length,

z_d hypothetical height at effective sink when the surface is assumed to work as perfect sink,

k Von Karmann constant (= 0.4),

k_c hypothetical gas-phase coefficient for mass transfer between air and plant phases,

u_* friction velocity,

L Monin-Obukhov length, and

ψ_c integrated form of universal function representing stability effect on mass transfer in the surface layer (e.g., Panofsky and Dutton, 1984)

The friction velocity u_* was estimated from,

$$u_* = \frac{ku\,(z_r)}{ln\left(\frac{z_r}{z_0}\right) - \psi_m\left(\frac{z_r}{L}\right)} \tag{9}$$

where ψ_m denotes the universal function in diabatic surface layer wind profile.
The value of z_0 was taken from Panofsky and Dutton (1984). The details of the resistances are described, for example, in Kitada and Ueda (1989).

(5) Treatment of Boundary Conditions

For horizontal boundary conditions, zero gradient of concentration was applied when wind blows toward outside of the domain, *i.e.* only advective mass flux is effective in this situation since this condition is introduced into calculation through "natural boundary condition" in FEM formulation. On the other hand, when air mass flows into the domain, inflow mass flux was set equal to advective flux with concentration at the boundary at the previous time step. The vertical boundary conditions are as follows:
at $z = H$,

$$K_v \frac{\partial C_i}{\partial z} = 0 \tag{10}$$

at $z = 0$,

$$K_v \frac{\partial C_i}{\partial z} = V_{g,i}\, C_i - \frac{Q_i}{\rho} \tag{11}$$

where H is the height of the modeling region and was 1.5 km above the ground, and Q_i is the emission source at surface level.

3.2 Model Application

(1) Description of Modeling Domain and Grid System

The modeling area, greater Dhaka (latitude 23°36′ to 24° N, longitude 90°9′ to 90°32′ E), covers 44 km in the north-south direction and 39 km in the east-west direction. The modeling area covers urban, suburban and rural area. A detail description about Dhaka is presented in section 2.2. The domain was chosen in such way so that the boundary conditions (concentrations) can be assumed to be small compared to the concentrations in the domain.

The modeling domain was discretized into 40 × 45 grid points with a spacing of 1 km in the horizontal direction, and in the vertical, the domain was divided into 17 layers with different thickness of 12, 18, 26, 34 m in the 1st, 2nd, 3rd, 4th layer, and 280 m at the uppermost model layer (17th layer), up to 1.5 km above the ground level.

(2) Initial and Background Concentrations

In the simulations, the initial and background concentrations of the chemical species can exert a significant influence on the model predictions. But due to the fact that the modeling domain was surrounded by relatively clean environments, as an extended domain was used in the simulation, inflow background concentrations were not a significant problem in this simulation. Initial concentrations, 1 ppb for SO_2, were assumed from the measured concentration

distributions. To minimize the impact of background concentrations on the model results, background concentrations were taken as initial concentrations.

(3) Meteorological Fields

Meteorological fields used in the model were prepared from the surface (10 m) data measured at every three hours at the Meteorology Observatory, Dhaka, during the study period. Because of nearly flat terrain in Greater Dhaka area and relatively narrow calculation domain, *i.e.*, 39 km × 44 km, we have assumed same wind speed at a height of 10 m above ground for the whole area as that at this meteorological observatory. The upper wind fields were extrapolated from the surface wind data using the log wind profile up to 100 m written as :

$$u = \frac{u_*}{k}\{ln\left(\frac{z}{z_0}\right) - \psi_m\left(\frac{z}{L}\right)\} \qquad z \geq z_0 \tag{12}$$

where u denotes the wind velocity at height z. The roughness length z_0=1 m for city center and 0.6 m for suburban and rural area were used in this study. Above the heights of 100 m, winds were set equal to those at 100 m height. As for the parameter related to thermal stratification in equations (6), (9), and (12), neutral condition was simply assumed.

Although we do not have data directly, showing surface heat flux, diurnal variation of the mixed layer similar to that over eastern India can be expected. Thus we used information about mixed layer in the eastern India.

In winter season the winds over Dhaka area can be characterized with their low velocity and persisting wind direction from north-west. They hardly show diurnal change in velocity and direction (see Figure 3 for example).

(4) Emissions Inventory

One of the most important parameters in air quality models is the emission inventory. Errors in the emission inventory cause air quality models to produce non-realistic results. These errors could propagate in a non-linear manner through the non-linear chemical processes.

The SO_2 and NO_x emission fields were prepared from the study of Azad and Kitada (1996). The emissions of non-methane hydrocarbon and carbon monoxide were derived from NO_x emission field. Emission of each non-methane hydrocarbon was specified in proportion to fuel-combustion-derived NO_x emission : in molar basis, $[NO_x]$: $[C_2H_6]$: $[C_3H_8]$: [ALKA] : $[C_2H_4]$: [ALKE] : [AROM] = 1 : 0.25 : 0.45 : 2.33 : 0.48 : 0.37 : 1.62, where $[C_3H_8]$ stands for propane and benzene, [ALKA] for lumped ≥ C_4 alkanes, [ALKE] for ≥ C_3 alkenes, and [AROM] for alkylbenzenes. The coefficients of proportion listed above were derived on the basis of the data for Aichi Prefecture, Japan (Nakanishi, 1996, and Nakanishi and Kitada, 1997). The emission sources were classified into two groups, the surface sources and the elevated sources. The surface sources included emissions from automobile, industry, residential activities, navigation vessel and commercial activities, while the elevated sources were brick fields. The effective stack height of 30 m (3rd vertical grid level) for brick fields was considered in this study. The brick fields are especially located in the north-west and south- east suburban of Dhaka (see Figure 2(b)). The inventories were compiled over a 39 × 44 km^2 region with 1 × 1 km^2 grid cells. The spatial distributions of total SO_2 emissions from surface and elevated sources are shown in Figure 5. The time dependence of the emission rates was described by the aid of assumptions for the diurnal variation of the major emission sources in Dhaka. The area sources reported were converted into the averaged point sources and then interpolated to the grid points.

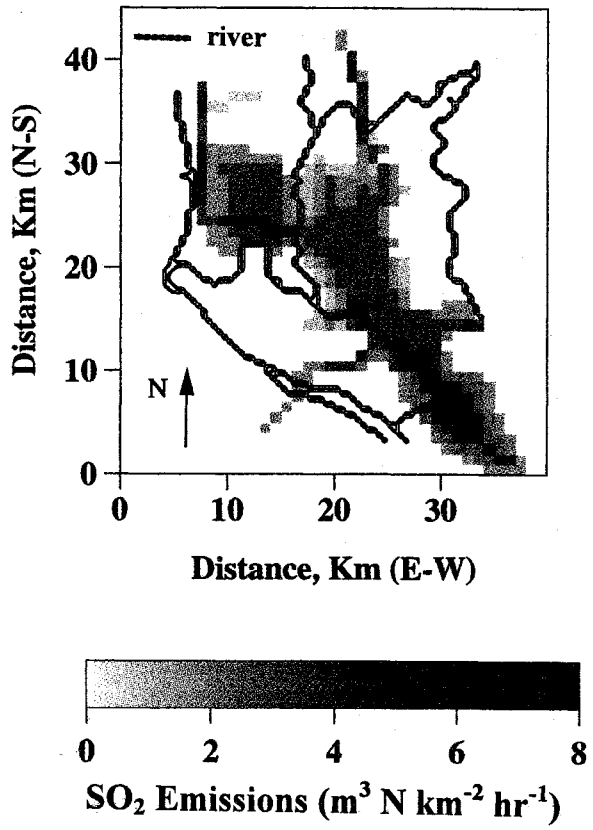


Figure 5. Spatial distributions of total SO_2 emissions from surface and elevated sources in Dhaka in winter 1995-96 (1 km × 1 km grid).

3.3 Model Results

(1) Calculated SO_2 Concentrations and their Comparison with Observation : Model Performance

Simulations were performed from 20 December 1995 to 30 December 1995 to compare calculated 10-day-average concentrations of SO_2 in Dhaka with those observed. The computed 10-day-average concentration distributions of SO_2, as shown in Figure 6, show that the south-eastern part of Dhaka, where brick field and other industry are located, exhibited the highest concentration of SO_2, and the city center also formed high concentration zone. The computed concentrations in Figure 6 indicate lower values in northern and western parts of the domain, reflecting the effect of predominant north-westerly on the pollutant transport. This characteristic distribution of the SO_2 concentration in Figure 6 agrees well with observation in Figure 4. This also suggests that long range transport of SO_2 discharged over India was not significant under weak wind; its direction ranging from north to west. Thus the SO_2 pollution in Dhaka in winter season is not affected by the Indian SO_2 emission since the prevailing wind in this season is northerly to westerly blowing from the Himalayas to the Bay of Bengal.

Model performance was evaluated by assessing the accuracy of the model predictions rel-

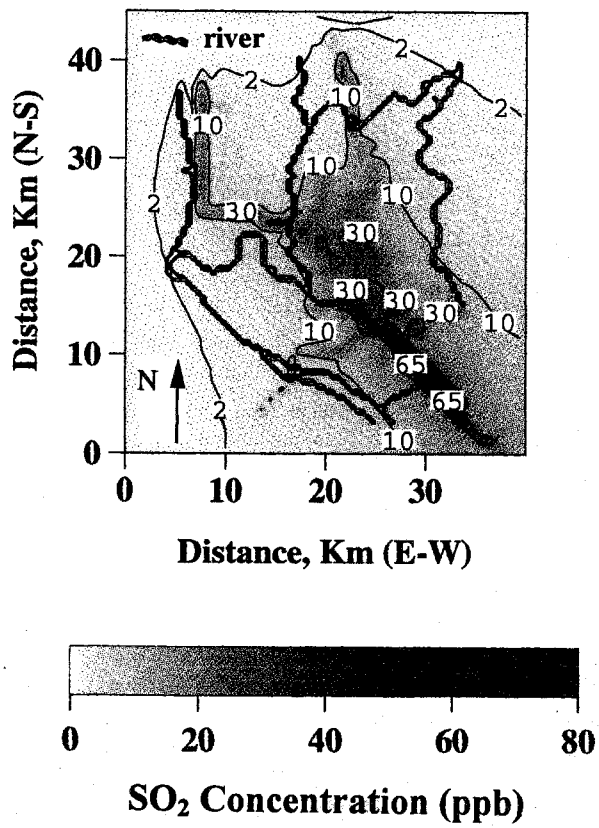


Figure 6. Computed 10-day-average concentrations of SO_2 in Dhaka in winter (20 to 30 December, 1995).

ative to observations. On the predicted ground-level SO_2 values, approximately 80% of the sites show agreement between modelled and observed concentrations to within $\pm 50\%$, and at about 50% of the sites the agreement is better than $\pm 25\%$. A comparison of the modelled and observed 10-day-average concentrations at 12 highly polluted sites in Dhaka is listed in Table 4, and it illustrates that the observed concentrations are well predicted by the model. In addition, we have calculated the correlation coefficient between model-predicted and observed concentrations for 64 sites data. The obtained high coefficient of 0.91 again demonstrates reliability of the simulation.

However, the model underpredicted the highest SO_2 concentration level in the south-east suburban area (see Table 4). This is most probably due to the lack of sufficient information regarding local emission sources. Among the other more likely reasons are inaccuracies in the interpolated wind fields and stack heights for point sources.

(2) Estimated Conversion Rate of SO_2 to SO_4^{2-} over Dhaka Area

The development of effective policies to control SO_4^{2-} deposition requires an understanding of how SO_4^{2-} is produced in the atmosphere. Based on the good agreement between computed and observed SO_2 concentrations (see subsection 3.3(1)), we have computed and analysed the

Table 4. Comparison of observed and calculated 10-day-average SO_2 concentrations at 12 highly polluted sites in Dhaka

Site	Observed Conc. (ppb)	Calculated Conc. (ppb)
Futulla	104	72
Pagla	78	71
Amin Bazar	46	38
Kachpur	43	44
Zinzira	43	41
Mohammadpur	43	42
Gabtali	41	37
Jatrabari	26	32
Dhaka-Aricha Road	39	37
Airport	16	21
Mothijheel	29	32
Farmgate	19	25

SO_4^{2-} to SO_2 ratio and its conversion rate in the atmosphere of Dhaka. In the simulations, the SO_x sources were splitted to those of SO_2 and SO_4^{2-} by 95 and 5% in volume basis respectively. To observe the SO_4^{2-} to SO_2 proportions in the atmosphere of Dhaka, we have calculated 3-day-averaged ground level SO_4^{2-} to (SO_2 + SO_4^{2-}) ratios, *i.e.*, the three days from December 20 to 22, 1995, and plotted those in Figure 7. The vertical profiles of SO_2 and SO_4^{2-} concentrations, and the ratio [SO_4^{2-} / (SO_2 + SO_4^{2-})] in the city center of Dhaka (Mothijheel) at noon on December 22, 1995 are shown in Figure 8. It is seen from Figure 7 that the 3-day-average ratio is relatively high in the peripheral area of Dhaka, which is located upwind in the prevailing north-westerly and has very little emission source. On the other hand, the ratio is relatively low in the emission source area such as south-east suburban and city center because of lower sulfate ratio of emission. The maximum and mean value of the 3-day-averaged ratio at ground level at Dhaka was 0.098 and 0.08 respectively. The increasing trend of the [SO_4^{2-} / (SO_2 + SO_4^{2-})] ratio in the vertical direction is seen from Figure 8. Above the height of daily mixing layer *i.e.*, 1.2 km, the ratio is very high at about 30%, indicating the situation that fresh SO_2 is not supplied from the below layer ; we have assumed uniform initial vertical profiles of SO_2 and SO_4^{2-} of 1 and 0.1 ppb, respectively.

In the model used here (Lurmann *et al.*, 1986), three chemical reactions are considered for the SO_2 to SO_4^{2-} conversion. We have estimated relative importance of those in the conversion mechanism. Those reactions are:

$$SO_2 + OH \xrightarrow{M,H_2O} SO_4^{2-} + HO_2 \quad (13)$$

$$SO_2 + CHO_2 \xrightarrow{M,H_2O} SO_4^{2-} + HCHO \quad (14)$$

$$SO_2 + CRO_2 \xrightarrow{M,H_2O} SO_4^{2-} + ALD_2 \quad (15)$$

where OH and HO_2 denote hydroxyl and hydroperoxyl radicals, CHO_2 and CRO_2 the CH_2O_2- and CH_3CHO_2- Criegee biradicals, respectively, ALD_2 the acetaldehyde, CH_3CHO, SO_4^{2-} the particulate sulfate and M the third body.

Using the spatial distributions of the 3-day-average value of K_{13}[OH] + K_{14}[CHO_2] + K_{15}[CRO_2] as the rate coefficients of pseudo first order chemical reaction, we calculated the

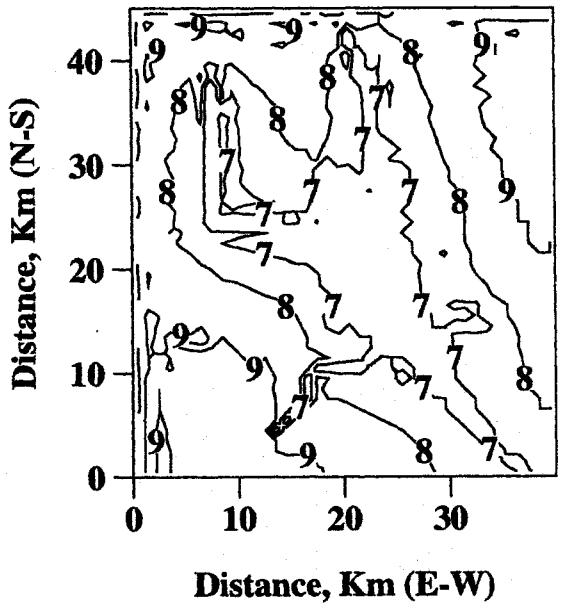


Figure 7. Spatial distributions of 3-day-average ground level $[SO_4^{2-} / (SO_2 + SO_4^{2-})]$ ratios (%) (starting from December 20, 1995) at Dhaka.

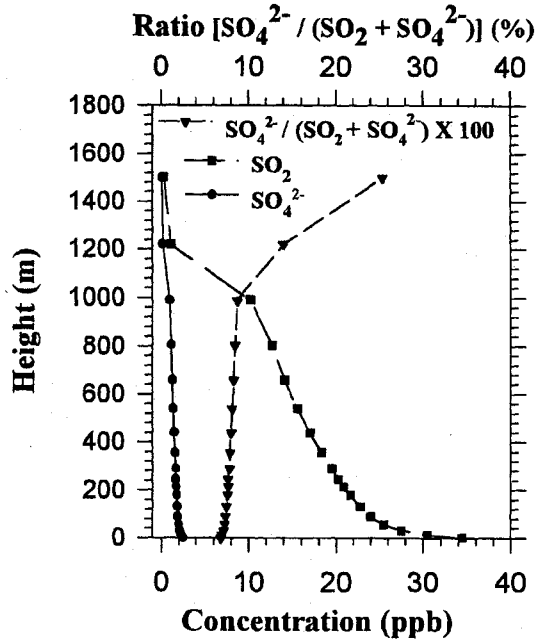


Figure 8. Vertical profiles of SO_2 and SO_4^{2-} concentration and the ratio $[SO_4^{2-} / (SO_2 + SO_4^{2-})]$ in the city center of Dhaka (Mothijheel) at noon of December 22, 1995.

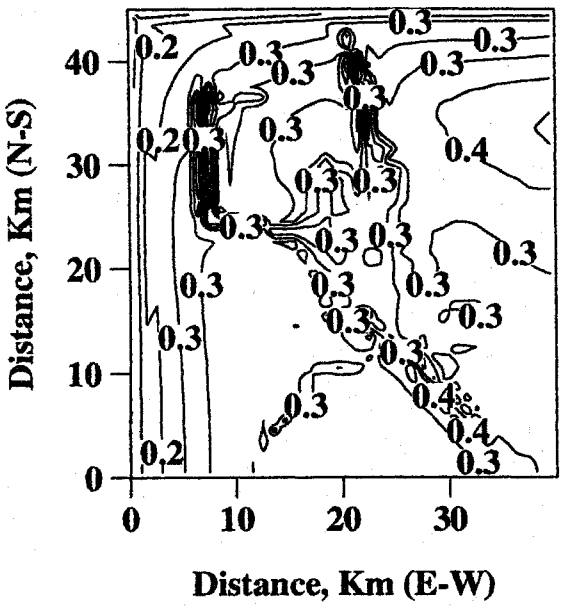


Figure 9. Spatial distributions of 3-day-average ground level conversion rates (% h^{-1}) of SO_2 to SO_4^{2-} at Dhaka (the calculation of conversion rates is discussed in the text).

spatial distributions of conversion rates of SO_2 to SO_4^{2-} in % $hour^{-1}$ at ground level as shown in Figure 9. The vertical profiles of the conversion rates of SO_2 to SO_4^{2-} at the city center of Dhaka at noon and mid-night on 22 Dec. are shown in Figures 10(a) and (b). The diurnal patterns of the total conversion rate and $[SO_4^{2-} / (SO_2 + SO_4^{2-})]$ ratio at ground level in the city center are shown in Figure 11. Figure 9 indicates high conversion rate at the emission source area, *i.e.*, south-east industrial zone and city center, and relatively low conversion rate at the area with almost no emission, *i.e.*, near north and west boundary. Figure 10 depicts increasing trends of conversion rate of SO_2 to SO_4^{2-} at mid- day (Figure 10(a)) and decreasing trends at mid-night (Figure 10(b)) in the vertical direction indicating the dependence of conversion rate on radical concentrations via day time solar radiation, which is reduced by high aerosol concentration near surface. The dense aerosol layer near the surface scatters and absorbs solar radiation which controls the photodissociation rates of chemical species. The present simulation used photodissociation rate coefficients for NO_2, O_3, HNO_3 etc., which were derived in Kitada and Peters (1980), based on the actinic flux estimated by Peterson (1976) by including higher aerosol concentration near surface in typical polluted area. The rapid increase of conversion rate at the mixing depth level (see Figure 10) is due to quick change of radical concentrations. The near surface level conversion rate in the city center of Dhaka (23°43′N latitude) at noon of December 22, 1995, which was 1.2% h^{-1} (see Figure 10(a)), is similar to 1% h^{-1} at mid-day of November at 25°N latitude in the study of Altshuller (1979). High value of conversion rate during day and low value at night is seen from Figure 11, indicating its dependence on radical concentrations. The diurnal patterns of $[SO_4^{2-} / (SO_2 + SO_4^{2-})]$ ratio is generally influenced by diurnal patterns of emission and conversion rate. The difference in the total conversion rate from day to day at noon (see Figure 11) is due to the changing concentrations of precursors of OH radical such as NO_x and non-methane hydrocarbons. These differences of concentrations from day to day were mainly caused by difference of wind speed, since other conditions such

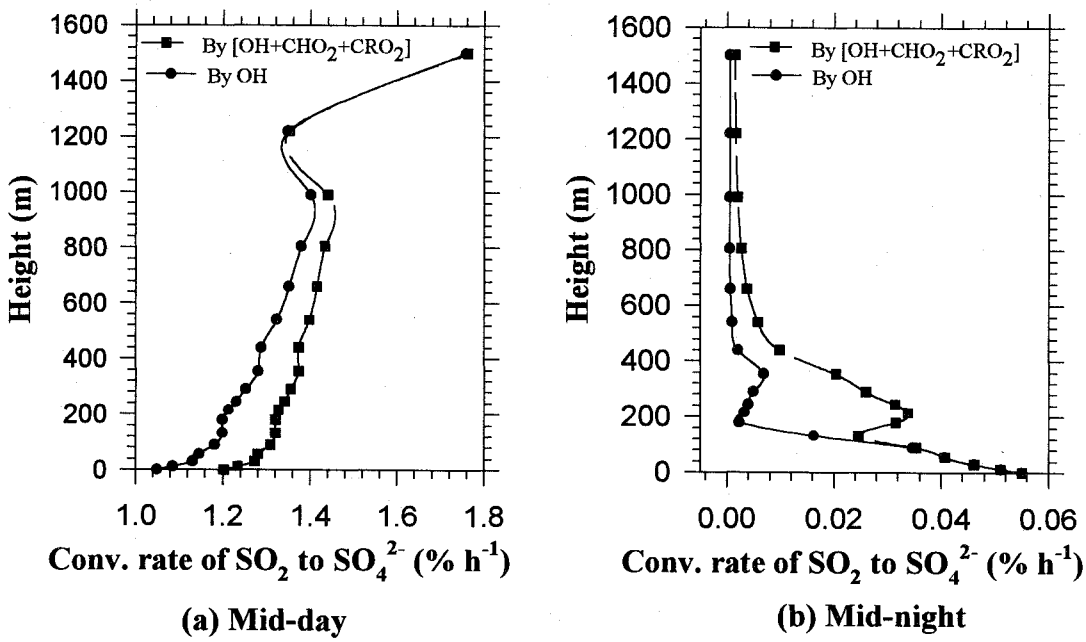


Figure 10. Vertical profiles of conversion rates (% h^{-1}) of SO_2 to SO_4^{2-} by OH and by $OH+CHO_2+CRO_2$ radicals at the city center of Dhaka at (a) noon, and (b) mid-night of December 22, 1995.

as solar radiation and activity of emission source were almost same.

The mean and maximum value of the 3-day-average total rate coefficient at ground level at Dhaka was 0.3 and 0.4 % h^{-1} respectively. Using this mean value as the rate coefficient of pseudo-first order chemical reaction the conversion rate of SO_2 to SO_4^{2-} near surface level in Dhaka was about 7% in a 24-hour period, which is larger than that reported in Eastern North America in the same season. The conversion rate in Eastern North America was within 3.1 - 4.7% in a 24-hour period in midwinter (Hidy, 1994). The higher conversion rate in Dhaka is most probably due to strong emission sources of NO_x and hydrocarbons in the city area, larger intensity of actinic flux and other meteorological parameter such as wind speed.

In this simulation, heterogeneous oxidation of SO_2 was not considered. Although there was no precipitation during the observation period and that is common in this dry season in this area, fog phenomena were observed near ground surface just before sunrise. Thus some aqueous phase oxidation possibly occurred within fog droplets. In the numerical study of the transport and chemistry of air pollutants under cloud formation condition over the Sea of Japan in winter, it is reported that oxidation rate of SO_2 was enhanced by about 25% on average over entire domain during 12 hours simulation (Kitada and Lee, 1993). Although physical and chemical situation in Dhaka is different from that over the Sea of Japan, we may assume that inclusion of heterogeneous reaction could increase the oxidation rate of SO_2 by 25% at most.

(3) Dry Deposition of SO_2

Based on the encouraging performance of the model to simulate SO_2 concentrations in Dhaka, we have calculated the dry deposition of SO_2 for Dhaka. The 10-days SO_2 deposition

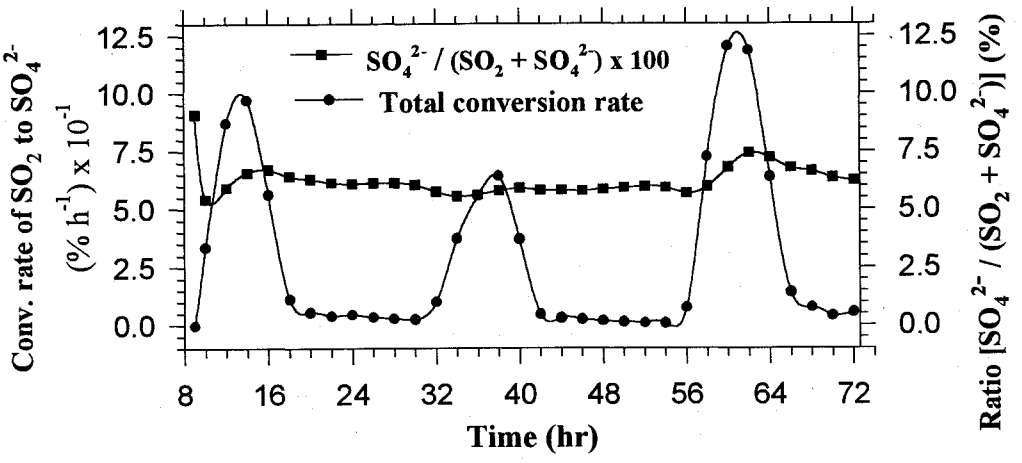


Figure 11. Diurnal patterns of the total conversion rate of SO_2 to SO_4^{2-} and [SO_4^{2-} / (SO_2 + SO_4^{2-})] ratio at ground level in the city center of Dhaka.

isopleths are shown in Figure 12. The units are kmol km^{-2} (10 days)$^{-1}$ as SO_2. The areas of largest deposition were south-east suburban and city center, and coincide with those of strong emission sources. The average and gross total deposition was 112.5 mol km^{-2} and 202.5 kmol as SO_2 respectively. No published data on Dhaka city are available for the comparison with the calculated dry deposition. The calculated mean dry deposition rate of SO_2 for Dhaka, 112.5 mol km^{-2} per 10 days, becomes about 0.97 kg SO_4^{2-} ha^{-1} for three winter months (December-February) after extrapolation. This value is higher to those reported for Agra, India and lower to those reported for Delhi, India in the same season. The sulfate deposition rate in Agra and Delhi for the cited three winter months was 0.54 and 1.95 kg ha^{-1} respectively (Sharma *et al.*, 1995).

However, the problems of acid rain have not yet been widely acknowledged in Bangladesh. The alkaline rich soil-derived particles in suspended particulate matter (SPM) are working in favour to neutralize acid rain in Dhaka. But, if the increasing trend of SO_2 in the atmosphere of Dhaka is continuing, the soil will lose its alkalinity and become acidic in near future.

(4) Sensitivity Analysis

Sensitivity analysis is useful both as a guide to the uncertainties involved in the model predictions, and to indicate which input parameter assumptions have the most influence on the model results. Sensitivity tests for three types of parameters, described below, were performed to investigate their effects on the predicted concentrations. The different cases for sensitivity tests are shown in Table 5.

(i)Initial concentration : The sensitivity tests for initial concentration included zeroing the initial SO_2 concentration (case 2), and setting initial SO_2 to 20 ppb (case 3) separately. It was observed that the effect of zeroing the initial concentration persisted only during the first 4 h of the simulation. The effects of taking 20 ppb as initial concentrations persisted upto 20 h of the simulation, indicating that short-term simulations with the model are sensitive to the initial concentrations, but multiday simulations would not be so sensitive. In the sensitivity tests for initial concentrations, the background concentrations were same to those at the base

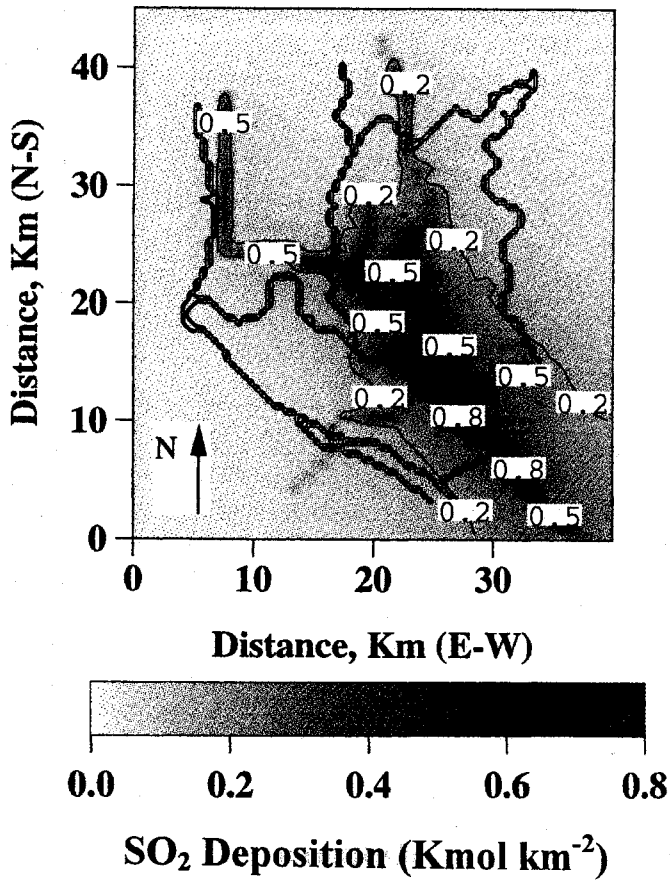


Figure 12. Computed 10 days' dry deposition of SO_2 (kmol km^{-2}) at Dhaka.

case simulation (1 ppb).

(ii) Roughness length : Two sensitivity tests for roughness length were performed: one case (case 4) increasing the roughness height by a factor of 2, and the other case (case 5) decreasing it by 50%. A surface roughness of 1 m for city center and 0.6 m for suburban area were used for base case simulation. In the case of the surface roughness increased by a factor of 2, the aerodynamic resistances to the mass transfer from atmosphere to surface decreased, resulting increase of dry deposition velocities, and thus the concentration of SO_2 decreased. For the case of surface roughness decreased, opposite results were observed. Both simulations began at 09:00 BST on December 20, 1995 with the same initial and boundary conditions as those in base case, and continued until noon of December 22, 1995. For the two days' simulations, doubling the roughness length (case 4) resulted in decrease of the mean and maximum SO_2 concentrations by 6.5 and 1.7%, respectively compared with the base case (case 1). Similarly, halving the roughness length (case 5) caused increase of those by 4.5 and 1.2%, respectively. The total deposition of SO_2 for the two days was increased by about 3.6% for doubling the roughness length and was decreased by 2.5% for 50% reduction of roughness length.

(iii) Mixing depth : In the simulations, an algebraic model for diurnal variation of mixing height was used (Eq. (3)), the model which was derived based on a boundary layer simulation

Table 5. Description of different sensitivity cases

Case No.	Initial Conc. (ppb)	Roughness Length (m)		Mixing Height
	SO_2	City Center	Suburban	
1 (Base case)	1	1	0.6	normal
2	0	1	0.6	normal
3	20	1	0.6	normal
4	1	2	1.2	normal
5	1	0.5	0.3	normal
6	1	1	0.6	doubling
7	1	1	0.6	halving

using $k - \epsilon$ turbulence model for hypothetical case, *i.e.*, on a sunny summer day over flat land surface in mid-latitude area. In the base case simulation diurnal variation of mixing depth was assumed with a maximum value of 1239 m at 15:30 BST and a minimum of 100 m from 23:00 to 06:30 BST. The mixing depth can vary from day to day as seen in the study of Gamo *et al.* (1994) on Indian sub- continent. To quantify the effects of mixing depth on the computed concentrations, two sensitivity tests were performed by increasing the mixing depth by 100% (case 6) and decreasing it by 50% (case 7). The simulations began at 09:00 BST on December 20, 1995 with the same initial and boundary conditions as those in base case, and continued until midnight of December 22, 1995. Due to doubling the mixing depth the maximum and mean ground level SO_2 concentration was decreased by 4% and 8.5% at noon, and by 19.5% and 22% at midnight of December 22, 1995. Reduction of mixing depth by 50% resulted in 12% and 28% increase in the ground level maximum and mean SO_2 concentration at noon of December 22, 1995 and 27% and 31% increase at midnight of the same day.

4. Conclusions

Ambient SO_2 concentrations have been measured in Dhaka city at 64 sites using diffusion tube samplers. An Eulerian transport/chemistry/deposition model has been also used to calculate SO_2 concentrations in Dhaka in winter. The model estimates were verified by measurements. An investigation on the chemical conversion of SO_2 to SO_4^{2-} is performed. Detailed sensitivity tests for three input parameters are also conducted. The overall results of the study can be summarised as follows :

(1) Dhaka city is highly polluted by SO_2. At some places SO_2 concentrations are more than two times to the Japanese ambient SO_2 concentration standard, which is 0.04 ppm for daily average of hourly values. So it is necessary to do realistic extensive sensitivity study for SO_2 emission reduction and find out optimised emission control system after cost-benefit analysis.

(2) SO_2 concentrations are high in the south-eastern industrial and brick field zone together with the route running from north-west to south-east, and also parallel to the Buriganga river.

(3) Good agreement was obtained between measurements and calculations. The correlation between modelled and observed concentrations was found to be 0.91. About 90% of the modelled values lay within a factor of 2 of the observed values.

(4) Estimated pseudo-first order chemical reaction coefficient of SO_2 to SO_4^{2-} conversion was 0.3 % h^{-1} (averaged value at surface level) for Dhaka area in winter.

(5) That the pseudo-first order reaction coefficient, which is approximately equal to the SO_2 to SO_4^{2-} conversion rate for 1 hour period, is not constant but shows diurnal- and spatial-variation which is clearly indicated in Figures 10 a and b (*i.e.*, its vertical profile for noon and mid-night) and Figure 11 (*i.e.*, diurnal variation at surface level).

(6) The sensitivity tests indicate that the model results are quite sensitive to the mixing height.

(7) The encouraging performance of the model indicates its suitability to investigate the effectiveness of emission reductions on the ambient pollutant concentrations.

The estimation of the contribution of various sources to the SO_2 distribution and the evaluation of source reduction strategies are the subjects of future studies.

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