



ENVIRONMENTAL STUDIES OF SO₂, SPM AND NO_x OVER AGRA, WITH VARIOUS METHODS OF TREATING CALMS

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Abstract—A comparative study between predicted and observed concentrations of major pollutants SO₂, suspended particulate matter (SPM) and NO_x in Agra, India has been undertaken using two Gaussian plume models. A Gaussian plume model (IITLT) and climatological dispersion model (CDM) have been used to estimate long-term concentrations of non-reactive pollutants due to emissions from area and point sources. The IITLT model is designed to treat the calm wind conditions ($u < 2 \text{ m s}^{-1}$). The computations show that about 10% of the total concentration of each type of pollutant at each station is due to calm winds alone. Therefore, consideration of calm winds is quite important. A comparative study of various methods of treating calm winds in IITLT model has been made. It is noted that in CDM model the central wind speed of the lowest wind speed class was arbitrarily taken as 1.5 m s^{-1} . This means that light winds or calm winds reported in the first wind speed class ($0-1.54 \text{ m s}^{-1}$) are assigned this value.

IITLT model has different dispersion parameters for moderate winds and calm winds, whereas CDM uses the same dispersion parameters for both the winds.

The monthly wind frequency tables and monthly mean concentrations of SO₂, SPM and NO_x for the period November 1975–March 1977 (Mathura Refinery Report, 1978) have been computed. The results obtained from the two models are found to be in good agreement with the observed values. A statistical error analysis of the observed and the predicted concentrations show a satisfactory performance of both models with RMSE of 1.38 for the IITLT model and 1.62 for the CDM. The value of index of agreement for IITLT model is 0.6 whereas for CDM it is 0.54.

Key word index: Emission inventory, dispersion model evaluation, calm wind condition, statistical analysis.

1. INTRODUCTION

Agra is a city of great historical importance where the Taj Mahal is located. The effect of environmental pollution on historical monuments in Agra city is a topic of vital concern. The setting up of the Mathura Refinery Complex, located 40 km upwind of Agra city, has highlighted the need for making a thorough study of the contributions of the refinery complex pollutants—sulphur dioxide (SO₂), suspended particulate matter (SPM) and oxides of nitrogen (NO_x) affecting the Taj Mahal and other historical monuments in Agra.

The common primary pollutants that cause material damage to construction materials like marble, sandstone and plaster are SO₂ and particulate soot. These primary pollutants are also indices of other pollutants emitted by the combustion of fossil fuel like coal, oil and other industrial activities. NO_x is a pollutant of secondary importance as it is due to high temperature combustion and undergoes photochemical reactions. Therefore, SO₂, SPM and NO_x (as NO₂) were selected as pollution parameters.

There is enough evidence to suggest that in recent years SO₂, SPM and NO_x levels in metropolitan cities of India have been increasing. Any new industrial siting powered by coal or fuel oil is likely to further degrade ambient air quality. In the context of our industrial development, the studies described below should be useful in decision-making processes, planning and management.

The major contributors of SO₂, SPM and NO_x (as NO₂) in Agra are domestic fuel consumers, industries, railway shunting yard and thermal power plants. The contribution from the Mathura Refinery adds to the emissions from local sources. It is, therefore, important to estimate SO₂, SPM and NO_x concentrations due to local sources in Agra to assist in developing protection measures.

In the present study, long-term concentrations of non-reactive pollutants due to local sources in Agra have been estimated by using two analytical dispersion models, IITLT and the climatological dispersion model (CDM). While the IITLT model has been developed at IIT Delhi, the CDM is an U.S. EPA guideline model (Adrian and Zimmerman, 1973). The

special feature of IITLT model is the treatment for the calm wind conditions through a puff diffusion approach.

2. MODEL CHARACTERISTICS

2.1. IIT long-term model (IITLT)

The receptor-oriented Gaussian plume model of Hanna (1974) has been adopted to obtain the concentrations of SO₂, SPM and NO_x due to area sources along with the monthly wind roses and the stability frequencies. The ground-level concentration at each receptor due to area sources is the sum of all the contributions of grids upwind of the receptor and is given by

$$C = \sum_j C_j = \frac{\sqrt{2/\pi}}{a(1-b)u} Q_j (X_j^{-a} - X_{j+1}^{-a}) \quad (1)$$

where C_j is the concentration due to an area source of strength Q_j located at the j th upwind grid, a and b are the stability parameters (Pasquill, 1974) given in Table 1a and u is the mean wind speed. X_j and X_{j+1} are the upwind distances of the j th and $(j+1)$ th grid from the receptor point.

Equation (1) requires that the grid system be aligned with the mean wind. By successive application of equation (1) to each wind direction sector, the model is able to give the monthly mean concentration of pollutants (SO₂, SPM and NO_x) at any receptor point after weighing it with appropriate combination of wind frequency, stability and speed class. Alignment of the source inventory grids with each of the wind direction is achieved in the manner given by Debberdt *et al.* (1973). For the elevated point sources a simple adoption of the basic dispersion equation due to Smith (1968) is used to calculate the concentration C_p at different receptor points:

$$C_p = \frac{360fQ}{\sqrt{2}\phi\pi^{3/2}u\sigma_{ze}} \exp\left(-\frac{h^2}{2\sigma_{ze}^2}\right) \quad (2)$$

where ϕ is the angular width of direction sector in degrees, f is the percentage frequency of occurrence of winds in a particular direction, wind group and stability during the period of interest, h is the effective stack height and σ_{ze} is the vertical dispersion parameter for an elevated source. Thus, the total concentration at any receptor point is the sum of C and C_p .

2.1.1. *Source inventory.* A gridded source inventory has been developed over an area of 12 km × 12 km as shown in Fig. 1. The importance of source strength in predicting SO₂, SPM and NO_x concentrations cannot be overemphasized. The most crucial parameters are the total city-wise hourly emissions of SO₂, SPM and NO_x. Ground-level area sources and elevated point sources have been considered separately. Major sources of SO₂, SPM and NO_x over Agra are the following.

Table 1a. Stability parameters and the eddy diffusivity coefficients

	a	b	a'	b'	K (m ² s ⁻¹)
Unstable	0.11	0.77	0.87	0.75	101
Neutral	0.05	0.68	0.66	0.64	101
Stable	0.03	0.65	0.23	0.58	5

(b)

Stability	A	B	C	D	$E \& F$
α (m s ⁻¹)	0.3	0.3	0.25	0.2	0.15
β (m s ⁻¹)	0.6	0.6	0.50	0.4	0.3

Source: Industrial Pollution Control (1989), General Review and Practice in Japan, Vol. 1, Air and Water, revised, Industrial Pollution Control Association of Japan, Tokyo, Japan.

- Industries include about 250 foundries around Agra. Most of these industries are situated to the northwest of the monuments. These foundries mostly use coal of the order of 200–300 tons per day.
- Railway shunting yard which is very close to Agra Fort. This marshalling yard uses approximately 40–50 tons of coal every day.
- Two power plants of 10 MW each. Approximately, one rake of coal (about 1100 tons) is used by these power plants daily.
- Domestic coal burning: Coal is still used for domestic cooking in Agra. The consumption of coal as domestic fuel is estimated as 24 g s⁻¹.

Emissions from (a), (b) and (c) are estimated from the average amount of coal consumed per day (Report-Expert Committee on Environmental Impact of Mathura Refinery, 1977); for (d), the annual consumption of coal in the entire area is used (U.P. Government Annual Statistics, 1981).

2.1.2. *Emission factors.* With sulphur content of Indian coal as 0.5%, the SO₂ emission factor is taken as 19S kg t⁻¹, where S is the sulphur content, the NO_x (as NO₂) emission factor is 3 kg t⁻¹ and that for suspended particulate matter is 10 kg t⁻¹. These emission factors for SO₂, SPM and NO_x have been obtained from the U.S. (EPA) report (Compilation of Air Pollutants Emission Factors, U.S. (EPA) 3rd edition, AP-42, Part-B).

Domestic sources have been apportioned over the entire area. Three sizes of square grids (0.25, 1 and 4 km² area) have been used so that more densely populated areas are treated in greater detail. The source strength S_i (in g s⁻¹) of the i th grid due to domestic coal burning is proportional to P_i the population in that grid. The grid sizes were chosen so that P_i is inversely proportional to a_i , the area of the i th grid. It follows that

$$S_i = k \frac{1}{a_i} \quad (3)$$

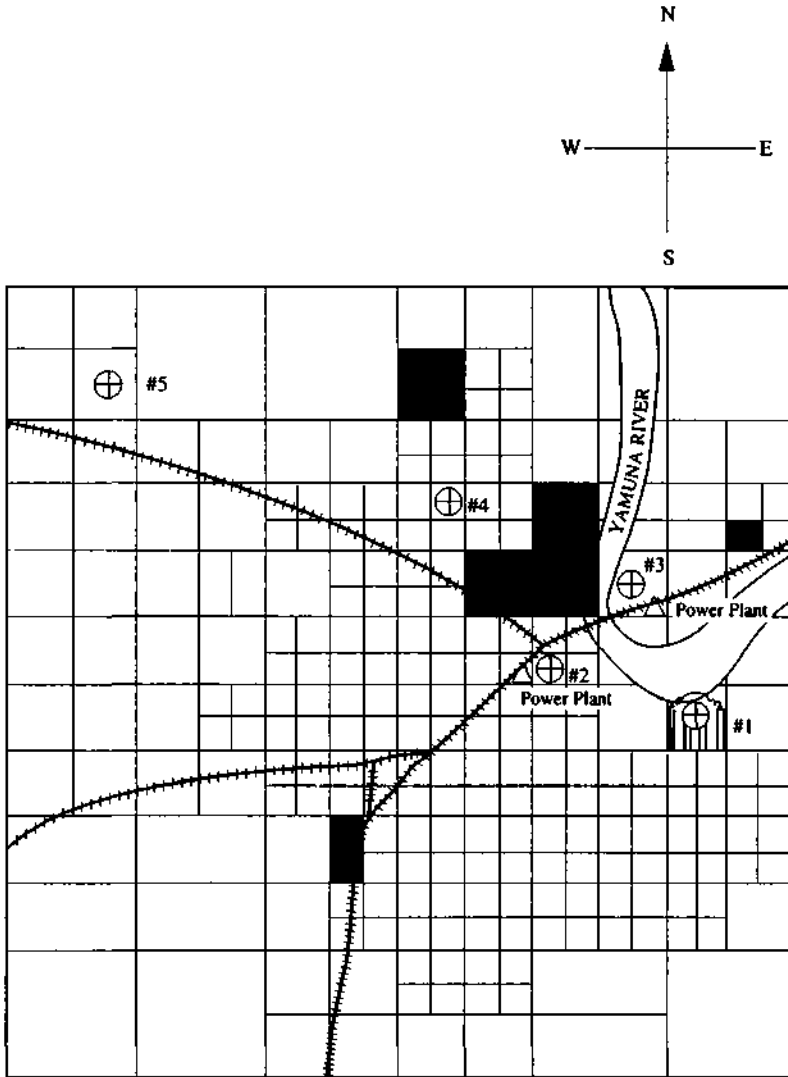


Fig. 1. Grid system over Agra and its surroundings. Slant lines show grids having industrial emission; cross-hatched area shows railway yard; (⊕) indicate concentration measuring station (receptors).

If n_1 , n_2 and n_3 are the number of grids and a_1 , a_2 and a_3 the corresponding areas in km^2 , then the total SO_2 emission S (in g s^{-1}), due to domestic coal burning is given by

$$S = \sum_i S_i = K \left(\frac{n_1}{a_1} + \frac{n_2}{a_2} + \frac{n_3}{a_3} \right). \quad (4)$$

The constant k in equations (3) and (4) can be estimated knowing S . In this way, the source strength in each grid has been apportioned. Depending on the locations of various industries and the railway shunting yard, the emissions from them are superimposed on these grids (Fig. 1).

2.1.3. Methods of treating calm winds in the IITLT model. One of the most frequently occurring meteorological conditions in India is the calm wind. In Agra, calms occur about 60% of the time. Calm wind effects are most significant in the vicinity of the sources. Pollutants emitted during the calm wind periods do

not travel or disperse readily from the source. Pollutants accumulate near the source till the whole cloud of pollutants are dispersed when wind speed increases. Long duration of calm conditions may also lead to an episodic condition. Gaussian models, in general, grossly overpredict concentration under low wind speed conditions. The present study deals with five different approaches for the treatment of calm wind conditions in the dispersion models.

Method 1: Dispersion of pollutants during calm wind conditions has been treated separately in the IITLT model by using an appropriate value of σ_z . For ground-based area sources, the model can be used with $\sigma_z = a'x^{b'}$, where a' and b' are the stability parameters computed for light wind conditions by using Smith's (1973) stability curves. Smith (1973) has numerically solved the steady-state advection-diffusion equation for unstable, neutral and stable cases for wind speeds of 3.0, 2.75 and 2.5 m s^{-1} at the 10 m level

corresponding to a geostrophic wind of 4 m s^{-1} . The values of parameters a' and b' have been estimated from these curves and are shown in Table 1a.

The ground-level concentration is a function of not only the dispersion parameters but also the mean wind velocity. The mean wind speed for calm winds in the IITLT model is taken as 0.5 m s^{-1} . Since the wind direction is variable under calm winds, it is assumed that all the directions are equally probable. Stability frequencies have been obtained from Pasquill's method (Turner, 1969).

For elevated point sources, we have adopted Draxler's (1979) approach which has defined σ_{ze} in terms of K , the coefficient of eddy diffusivity for each Pasquill stability category. Thus,

$$\sigma_{ze} = \sqrt{2K \frac{x}{u}}$$

Table 1a lists the values of K for different stabilities, which are used in this work.

Method II: In hourly climatological calculations the calm wind is apportioned into the next non-calm wind direction that occurs. Here, calm wind speed is taken as 1 m s^{-1} . Values of dispersion parameters are the same as in the preceding method.

Method III: This method of treatment of calm winds in the IITLT model is the same as the one adopted for calm winds in the CDM. Calm wind frequency is assumed to follow the same distribution as the lowest wind speed category. Wind speed for calm is assumed as 1 m s^{-1} . Dispersion parameters have been estimated from Pasquill-Gifford (P-G) curves.

Method IV: Another method for handling calm winds is to use a series of puffs. Csanady (1973) suggested the use of integrating a series of instantaneous puffs released for the period of calm.

For calm conditions the "puff" formulation used in the present work for estimating the ground-level concentrations is given as

$$C = \frac{q}{(2\pi)^{3/2} x^2 \beta R^2} \exp\left(-\frac{R^2}{2t^2}\right), \quad (5)$$

where q is the emission rate (g s^{-1}), u is the mean wind speed (m s^{-1}), H_e is the effective stack height (m), t is the diffusing time (s), $R^2 = ((x^2 + y^2)/x^2 + H_e^2/\beta^2)$, $\sigma_y = xt$ and $\sigma_z = \beta t$, where σ_y , σ_z represent the plume growth along the y and z axis, respectively (m), and α and β depend on the stability conditions of the atmosphere (Table 1b). Equation (5) has been taken from a book entitled "Industrial Pollution Control Association" (1989). It is integrated over a time period w.r.t. diffusion time t for concentrations. The integral form of equation (5) is given by

$$C = \frac{q}{(2\pi)^{3/2}} \int_0^t \frac{1}{\sigma_y^2 \sigma_z} \exp\left[-\frac{1}{2} \left(\frac{x^2 + y^2}{\sigma_y^2}\right)\right]$$

$$\exp\left[-\frac{1}{2} \frac{H_e^2}{\sigma_z^2}\right] dt. \quad (6a)$$

Equation (6a) can be written as

$$C = \frac{q}{(2\pi)^{3/2}} \sum_i \frac{1}{(xt)^2 \beta t} \exp\left[-\frac{1}{2} \left(\frac{x^2 + y^2}{(xt)^2}\right)\right] \exp\left[-\frac{1}{2} \frac{H_e^2}{(\beta t)^2}\right] \Delta t. \quad (6b)$$

In order to calculate the concentrations, the persistence time of 2 h is assumed and a time interval of 10 s is taken for the puff to reach the receptor. In order to take into account the reflection from the ground the term $\exp(-H_e^2/2\sigma_z^2)$ is modified to

$$\exp\left(-\frac{H_e^2}{2\sigma_z^2}\right) + \exp\left[-\frac{(H_e - 2L)^2}{2\sigma_z^2}\right].$$

Hence equation (5) is given by

$$C = \frac{q}{(2\pi)^{3/2} x^2 \beta R^2} \exp\left[-\left(\frac{x^2 + y^2}{2\sigma_y^2}\right)\right] \left[\exp\left(-\frac{H_e^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(H_e - 2L)^2}{2\sigma_z^2}\right) \right]. \quad (7)$$

Equation (7) has been used for elevated point sources under calm conditions. In this equation, diffusion from the top has been limited by mixing depth, L . When σ_z becomes larger than eight-tenths of L , σ_z is replaced by L (Singh *et al.*, 1990).

During the computations of ground-level concentrations from puff formulations from equations (6a) and (7), different steps adopted are summarized below.

(i) Calculation of concentrations for any one source for different time intervals (namely $t = 10, 20, 30$ min, etc.).

Check

$$[C_i(t = 10n)/C_i(t = 10(n-1))] < 0.1, n = 2, 3, \dots$$

Stop computations for the lowest n (say $n = 3$) satisfying the above inequality and add concentration upto the time step ($t = 10n$).

(ii) Similarly, the same steps have been followed for other sources.

(iii) Once the addition of concentration for different time steps (for any one source) is done, check has been made on $[C_{i+1}(t)/C_i(t)] < 0.1$. If it is true then C_{i+2} onwards have been neglected.

Finally, the concentration, for moderate winds ($\geq 2.0 \text{ m s}^{-1}$) and calm winds ($< 2.0 \text{ m s}^{-1}$) are, respectively, calculated at different receptor points from equations (1), (2) and (5), (7). It is weighed by the corresponding frequencies of moderate and calm winds in the following fashion:

$$\text{(total concentration)} C = \sum C_1 (\text{moderate}) \times f_w + \sum C_2 (\text{calm}) \times f_c$$

where C_1 is the concentration for moderate winds obtained from equations (1) and (2) and f_w is the

monthly wind frequency for winds $\geq 2.0 \text{ m s}^{-1}$, C_2 is the concentrations during calm wind conditions which is estimated from equations (5) and (7) and f_c is the calm wind frequency for one month.

In this way, monthly mean concentration of pollutants has been obtained.

Method V: This approach, incorporating series of puffs to simulate dispersion pattern under calm conditions, is based on Deardorff's work (1984).

Estimation of the ground-level concentration is made for a point source, by puff diffusion in a thoroughly mixed convective boundary layer, under calm winds ($u \leq 2.0 \text{ m s}^{-1}$). This study incorporates a continuous succession of the mean circular puffs emanating from a point source, with each spreading puff having a standard deviation σ_r , represented by (i) and travel time t .

$$\sigma_{R_0}^2 = \left(\frac{\sigma_r}{Z_i}\right)^2 = \frac{(0.6T)^2}{(1+2T)} + \sigma_{R_0}^2 \quad (\text{i})$$

where σ_{R_0} is the initial spread of the puff at $t = 0$ and $T = (w_*/Z_i)t$, where w_* is the convective velocity scale and Z_i is the boundary layer height. This model assumes a Gaussian distribution radially within each mean puff and, hence, does not treat concentration fluctuations.

Under calm wind conditions, the ground-level concentration can be estimated from the Gaussian puff model given by

$$C = \frac{Q}{(2\pi)^{3/2} \sigma_x \sigma_y \sigma_z} \exp\left(-\frac{r^2}{2\sigma_z^2}\right) \times \left[\exp\left(-\frac{(z+H)^2}{2\sigma_x^2}\right) + \exp\left(-\frac{(z-H)^2}{2\sigma_x^2}\right) \right]$$

It is assumed in the above equation that $\sigma_x = \sigma_y$, expressed as σ_r , where $r = (x - ut)^2 + y^2$.

When σ_z becomes larger than eight-tenths of the mixing height, Z_i , the puff is assumed to be well mixed and concentration is expressed as

$$C = \frac{Q}{2\pi\sigma_x^2\sigma_z} \exp\left[-\frac{r^2}{2\sigma_z^2}\right] \quad \text{for } \sigma_z > 0.8Z_i. \quad (\text{ii})$$

Now consider a single puff emanating from a point source and well mixed within the convective boundary layer ($0 < z < Z_i$). Thus, the mean incremental concentration dC , a function of time, t' is given by

$$dC = \frac{dM}{2\pi\sigma_r^2(t')Z_i} \exp\left[-\frac{r^2}{2\sigma_r^2(t')}\right] \quad (\text{iii})$$

where dM is the mass of the net contaminant.

The mean concentration \bar{C} for a continuous source, can be obtained by integrating (iii) over time t :

$$\bar{C}(t) = \int_0^t dC = \int_0^t \frac{Q}{2\pi Z_i \sigma_r^2(t')} \exp\left[-\frac{r^2}{2\sigma_r^2(t')}\right] dt' \quad (\text{iv})$$

where $dM/dt = Q$ is the source emission rate (expressed in g s^{-1}).

By using the mixed layer scaling with w_* as the only velocity scale, the concentration can be written as

$$C_* = \bar{C} Z_i^2 \frac{w_*}{Q} = (2\pi)^{-1} \int_0^T \sigma_{R_0}^{-2}(T') \exp\left(-\frac{R^2}{2\sigma_{R_0}^2(T')}\right) dT' \quad (\text{v})$$

where

$$R^2 = \frac{x^2 + y^2}{Z_i^2} = \frac{r^2}{Z_i^2} \quad \text{and} \quad T' = \frac{w_* t'}{Z_i}$$

Finally, x is replaced by $x - ut'$ in equation (v) to consider the effect of the mean wind along x .

$$C_* \left(\frac{x}{Z_i}, \frac{y}{Z_i}, T, \frac{u}{w_*} \right) = \frac{1}{2\pi} \int_0^T \sigma_{R_0}^{-2} \exp\left[-\frac{\left(\frac{x}{Z_i} - \frac{uT'}{w_*} \right)^2 + \left(\frac{y}{Z_i} \right)^2}{2\sigma_{R_0}^2(T')} \right] dT' \quad (\text{vi})$$

The integral in equation (vi) is solved numerically for a period of 1 h.

The model evaluation is done for calm winds with wind velocity $u = 0.5 \text{ m s}^{-1}$. The convective velocity is taken to be $w_* = 1.8 \text{ m s}^{-1}$. The boundary layer height (mixing height) in the well-mixed convective boundary layer is considered 1500 m and the initial spread of the puff, σ_{R_0} , is taken to be $\sigma_{z_0}/Z_i = 0.005$.

The rest of the computations (to obtain the concentrations) follow the procedure outlined in method IV except that equation (vi) is used [instead of equations (5) and (7)] for calm winds.

The percentage of occurrence of all stability classes have been compiled separately using Turner's (1969) table. Monthly mean mixing heights for Delhi computed by Manju Kumari (1985) are directly used in the present work. Monthly mean mixing heights under different stability conditions of the atmosphere for 1 yr have been shown in Table 2.

2.1.4. Incorporation of dry deposition of SO_2 . Most of the atmospheric pollutants are eventually removed from the atmosphere by deposition on vegetation, soil or water. Methods of transport to vegetation, soil or water include dry deposition and precipitation scavenging. In the present study, we have included dry deposition for SO_2 .

Very small particles and gases are also deposited on surfaces as a result of turbulent diffusion and Brownian motion. In this case, a deposition velocity, V_d , can be defined as an empirical function of the observed deposition rate, w , in mass per unit area per unit time and concentration C_0 near the surface (Hanna *et al.*, 1982), i.e. $V_d = w/C_0$ (m s^{-1}).

The height at which C_0 is measured is typically about 1 m. Once V_d is known for a given set of conditions the formula $w = V_d C_0$ can be used to predict dry deposition of gases and small particles where

Table 2. Mixing heights (m) of 12 different months [January–December (1968–1972)]

Type of stability	January and February	March and April	May and June	July and August	September and October	November and December
A	1500	2000	2200	1800	2000	1500
B	1000	1500	1500	1500	1800	1200
C	1000	1500	1500	1500	1500	1000
D	1000	1200	1000	1000	1000	1000
E	350	550	1000	1000	500	350
F	100	350	500	500	350	100

Table 3. Definition and units

Symbol	Definition	Units
F	Buoyancy flux parameter	$\text{m}^4 \text{s}^{-3}$
g	Acceleration due to gravity	m s^{-2}
H	Effective height of plume	m
h'	Stack height adjusted for stack downwash	m
s	Stability parameter	s^{-2}
T	Ambient air temperature	K
$u(h)$	Wind speed at stack top	m s^{-1}
X_f	Distance to final rise	m
X^*	Distance at which atmospheric turbulence begins to dominate entrainment	m

C_0 can be obtained from some appropriate diffusion model. In the present model C_0 has been calculated from IITLT model.

A suitable deposition velocity for SO_2 was obtained from an extensive survey of the literature (McMohan and Dansion, 1979). The value chosen was 0.01 m s^{-1} . Table 5 shows the SO_2 deposition rate at different locations over Agra.

In our computation for concentration, we have used the approach adopted in the CDM for dry deposition by introducing the exponentially decaying factor $\exp(-0.0692 \rho/U_1 T_{1/2})$, where ρ is the distance between source and receptor, U_1 is the representative wind speed and, $T_{1/2}$ is the assumed half-life of the pollutant (h). In the case of SO_2 , half-life is 4 d (Hanson and Seller, 1984); the same decay expression has been used in the climatological dispersion model.

2.1.5. *Plume rise.* Briggs' (1969, 1971, 1973, 1975) plume rise formulae for hot plumes are used in both the models (IITLT and CDM) for evaluating concentrations from elevated point sources. These plume rise formulae are summarized below. For unstable or neutral atmospheric conditions, the downwind distance of final plume rise is $X_f = 3.5X^*$, where $X^* = 14F^{5/8}$ for $F < 55 \text{ m}^4 \text{ s}^{-3}$, and $X^* = 13F^{2/5}$ for $F > 55 \text{ m}^4 \text{ s}^{-3}$.

The effective stack height under these conditions is

$$H = h' + [1.6F^{1/3}(3.5X^*)^{2/3}/u(h)]. \quad (8)$$

For stable atmospheric conditions, the downwind distance of final plume rise is

$$x_f = 0.0020715u(h) s^{-1/2}$$

where $s = g(\partial\theta/\partial z)/T$. The effective stack height is given by

$$H = h' + 2.6\{F/[u(h)s]\}^{1/3} \quad \text{for windy conditions} \quad (9)$$

$$H = h' + 4F^{1/4}s^{-3/8} \quad \text{for near calm conditions.} \quad (10)$$

The lesser of the two values obtained from equations (9) and (10) is taken as the final effective emission height. Definition and units of symbols used in the plume rise equations are given in Table 3.

It is assumed that the source is located at the centre of each grid for area sources without any consideration of source density and choice of grid size in the relevant region. The IITLT model (improved version with method V) and CDM have been used to calculate concentrations of SO_2 , SPM and NO_x in each of the 16 wind directions at the five receptor points in Agra (Fig. 1).

2.2. Climatological Dispersion Model (CDM)

CDM, a U.S. operational model, is used in estimating long-term concentrations of non-reactive pollutants due to emissions from area and point sources in

the urban area. Two pollutants may be considered simultaneously in this model. This model is available as part of UNAMAP (version 6) and model formulations are presented in user's Guide (EPA-R4-73-024; December 1973, Environmental Monitoring Series).

2.2.1. *Calm winds in CDM.* Calm winds (low winds) have not been treated separately in the CDM. The central wind speed of the lowest wind speed class was arbitrarily taken as 1.5 m s^{-1} . This means that light winds reported in the first wind speed class are rounded up to this value. Operational wind instruments are generally designed for durability and also to withstand exposure to strong, gusty air flow and so most of the operational wind instruments do not sense low wind speeds accurately. For these reasons most wind sensors have a high starting speed and wind directions are very much suspected, which can lead to erroneous reporting of light winds as calms (Truppi, 1968).

2.2.2. *Model evaluation.* The computer program to solve equations has been developed at IIT Delhi to calculate monthly mean of SO_2 , SPM and NO_x concentrations for the city of Agra.

The required input parameters are as follows:

- (1) Source strength of each type of source.
- (2) Locations of the receptors at which concentrations are required.
- (3) Meteorological data: monthly wind roses, stability frequencies for different stabilities, mixing height for each month.

(4) Stack characteristics for elevated sources. Meteorological parameters. The monthly wind frequency tables for the period November 1975–March 1977 were compiled from the following two sources:

- (a) The National Environmental Engineering Research Institute (Mathura Refinery Report, 1978).
- (b) Environmental Impact of Mathura Refinery-Observational Programme For Investigation of Pollution, Interim Report No. 2 by India Meteorological Department (1979).

3. RESULTS AND DISCUSSION

A computer program of IITLT model, installed at the ICL 2960 computer system, is further improved by introducing different methods for treating calm wind conditions. Table 4 shows the output from different methods of calm winds in IITLT model and the CDM as well as observed concentrations of SO_2 , SPM and NO_x at Taj Mahal, Agra Fort, Itmat-ul-Daulah, Nagar Maha Palika and Sikandra in Agra. It has been shown in Table 4 that ground-level concentrations, computed from the IITLT model with method V for calm winds are closer to the observed levels of SO_2 , SPM and NO_x in comparison to the results obtained by using methods I, II and III for calm winds in the IITLT model. Methods I and II give the results which

Table 4. Monthly mean (November 1975–March 1977) concentrations (in $\mu\text{g m}^{-3}$) of SO_2 , SPM and NO_x due to area and point sources including calm wind conditions at Agra

S. No.	Pollutant	IITLT model prediction					Without calm wind	CDM predictions and Method III	Observed concentration
		Method I	Method II	Method IV	Method V				
Taj Mahal									
1.	SO_2	19.8	20.1	26.9	21.8	17.4	27.3	13.9	
2.	SPM	347.9	313.2	442.0	399.9	308.7	442.5	254.3	
3.	NO_x	22.8	11.2	27.1	25.0	20.4	27.0	14.8	
Agra Fort									
1.	SO_2	29.9	18.0	33.4	33.4	26.2	35.9	19.6	
2.	SPM	438.9	288.0	501.2	489.0	390.2	502.8	305.2	
3.	NO_x	27.3	11.2	31.0	29.2	22.6	31.9	17.7	
Itmat-ul-Daulah									
1.	SO_2	28.2	12.9	37.9	32.4	25.0	38.4	24.0	
2.	SPM	386.0	690.0	499.0	422.0	330.7	499.9	325.5	
3.	NO_x	24.2	14.7	32.0	27.0	21.0	33.0	17.4	
Nagar Maha Palika									
1.	SO_2	18.9	17.7	17.9	22.8	17.8	18.4	14.2	
2.	SPM	310.0	410.6	392.0	347.0	270.0	392.5	297.9	
3.	NO_x	20.9	20.1	26.9	23.8	18.9	27.4	18.2	
Sikandra									
1.	SO_2	9.2	11.3	11.0	9.9	7.2	11.4	8.8	
2.	SPM	185.8	305.0	233.0	209.7	165.0	233.7	186.1	
3.	NO_x	9.7	10.3	11.9	9.8	8.0	12.7	9.1	

are close to each other, because, in both the methods, Smith's (1968) curves have been used for the computations of dispersion parameters. These two methods are different in the choice of frequency distribution of 16 wind directions under calm winds. Method IV (Puff formulation) which is quite independent of these two methods does not have any assumption about frequency distribution of calm winds. The only assumption which is made in this method is the persistence time of 2 h. The concentrations obtained from IITLT model are still on the lower side compared to the results obtained from the CDM. The results of the IITLT model with method III for calm winds and the CDM are almost the same. On the basis of comparison between model-predicted values and observed values, the IITLT model with method V for calm winds seems to be quite appropriate for treating calm winds in Gaussian plume formulation. The comparison of results of the CDM, observations, and the IITLT model with method V for calm winds have been made in Table 4 which shows that the IITLT model and the CDM are comparable but both overpredict in comparison to the observed concentrations. The IITLT model results are close to observed values in comparison to the computations from the CDM. It should be noted that, in general, there is a tendency for the models to overpredict the SO_2 , SPM and NO_x levels. According to Hanna (1982), the criterion for assessing the performance of the model is that the natural variability of the deviations should be within a factor of 2-3 of the actual values. Predicted concentration of both the models are within a factor of 2 of the observed concentrations.

The present calculations indicate that the maximum ground-level concentrations occur in each month for the northwesterly and westerly winds at all the receptor points. This is consistent with the wind roses prepared by NEERI at Agra and with the presence of foundries located in NW direction of the city.

Table 4 shows that about 10% of the total concentration of each pollutant at each station is due to calm winds alone. Therefore, consideration of calm winds for cities like Agra is quite important.

Table 4 further indicates that monthly mean concentration of SO_2 is around $20 \mu\text{g m}^{-3}$ at most of the stations except Agra Fort and Itmat-ul-Daulah. At these stations concentration levels reach $30 \mu\text{g m}^{-3}$. The reason may be that both the power plants are also located very close to Agra Fort and Itmat-ul-Daulah. At Sikandra, SO_2 concentration level is quite low, since it is situated at the edge of the city. In Agra, the most important monument is Taj Mahal. Figures 2-4 show the comparative monthly mean concentrations computed from the IITLT model and the CDM as well as the observed concentrations of SO_2 , NO_x and SPM at Taj Mahal. The air quality standards laid down by the Government of India under the Air Prevention and Control of Pollution Act 1981 (Annual Report 1982-83) for the quality of air concerning SO_2 is $30 \mu\text{g m}^{-3}$ as an annual average. This standard

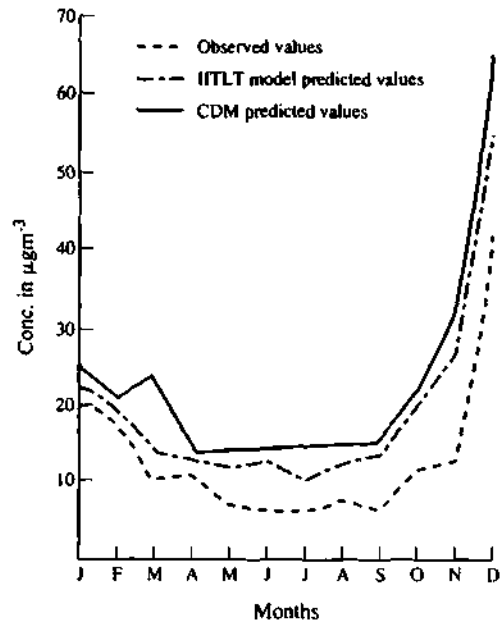


Fig. 2. Monthly averages of SO_2 concentration in ($\mu\text{g m}^{-3}$) at Taj Mahal (Agra).

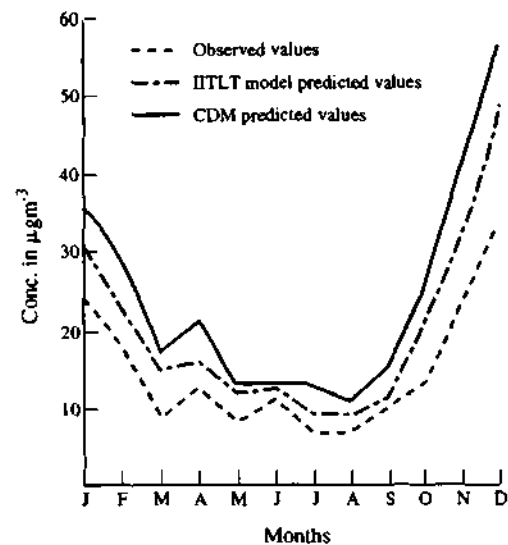


Fig. 3. Monthly averages of NO_x concentration in ($\mu\text{g m}^{-3}$) at Taj Mahal (Agra).

governs sensitive areas including tourist resorts, sanctuaries and national monuments, etc. Figure 2 shows that at Taj Mahal, SO_2 levels never exceed air quality standards. Even though the total amount of emission of SO_2 from local sources may be small, on account of their proximity to the monuments, their contribution to the air quality of the zone near the monuments will be considerably high. Most of the time, NO_x concentrations at different stations, were found to be close to the lowest detectable level ($20 \mu\text{g m}^{-3}$). It may, therefore, be concluded that the NO_x impact is negligible (Ref. Baseline Air Quality Study at Agra. Second

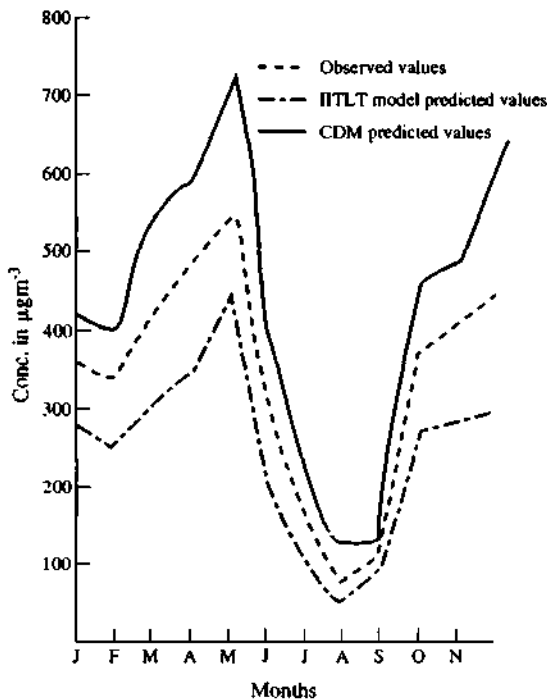


Fig. 4. Monthly averages of SPM concentration in ($\mu\text{g m}^{-3}$) at Taj Mahal (Agra).

Quarterly Report, 1976 summer data, Report by NEERI, Nagpur). The result of monthly mean computations for SPM ranges from 100 to 500 $\mu\text{g m}^{-3}$. Higher values during April and May can be attributed to the stormy and dusty weather which prevails in this area. In these two months, predicted values exceed the Indian standard (100 $\mu\text{g m}^{-3}$).

In the present study, dry deposition of SO_2 has been computed and shown in Table 5 at different receptor points in Agra. The possibility of pollutant (SO_2) removal by physical or chemical processes is included in the program by introducing the decay expression $\exp(-0.692\rho/U_1 T_{1.2})$. The chemical conversion of SO_2 to SO_4^{2-} has not been considered in the present work.

Statistical analysis of the predictions and the observations is central to the model performance evaluation. The methods discussed by Fox (1981) and Rao *et al.* (1985) are used for model evaluation and comparison with the CDM. The root mean square error

(RMSE) was determined for each pair of calculated and observed values.

$$\text{RMSE} = \exp \left[\frac{1}{n} \sum_i (\ln C_p - \ln C_o)^2 \right]^{1/2}$$

where n is the total number of pairs of predicted (C_p) and observed (C_o) concentrations. Assuming that the distribution of observed to calculated concentration ratios is normal, RMSE is the standard deviation of this ratio. Thus, a perfect model with complete agreement between observed and predicted concentrations would have RMSE of 1 (Raghavan *et al.*, 1983). The 170 (17 months \times 5 stations \times 2 models) pairs of predicted and observed concentration at five stations give RMSE of 1.38 for IITLT model and 1.62 for CDM which show that both the models perform satisfactorily.

The overall measure of bias for both the models (-1.31 for IITLT and -1.67 for CDM) shows negative values which reflect the fact that both the models have a tendency to overpredict. This is probably due to the fact that we have adopted Pasquill's scheme of stability classification and the empirical form of dispersion formulation $\sigma_z = ax^b$. Besides, it is well known that Gaussian formulation tends to overpredict.

Regarding the emission inventory, it is well known that area sources are difficult to estimate as compared to point sources (Ku *et al.*, 1987). The correct characterization of a source as point or area is also important for the proper simulation of the concentration field in an urban area. Usually, the models treat the area sources as ground-level sources and no plume rise formulation is applied. The importance of the area sources (e.g. spatial heating) during the winter may be one of the reasons for larger error in the winter case.

Area sources dominate during the nighttime stable period while the point sources dominate during the daytime convective conditions. During the nighttime, the low mixing layer depth prevents the emissions from the elevated point sources from diffusing to the surface and low mixing layer as well as small eddy diffusivity prevents the area source emissions from mixing in the vertical. This causes the area source emissions to dominate the concentration distribution during the night. Convective turbulence breaks up the surface-based inversion and the fumigation process leads to an increased contribution from point sources. In contrast, the increase of mixing depth and eddy diffusivity reduce the contribution from area sources.

Another descriptive statistic is the index of agreement (Willmott, 1982) which is a measure of the degree to which the observed variate is accurately estimated by the simulated variate. This is not a measure of correlation or association in the formal sense, but rather a measure of the degree to which model predictions are error-free. At the same time it is a standardized measure so that cross comparisons of its magnitude for different models can be made. The

Table 5. Rate of removal of material (SO_2) due to dry deposition effect

S.No.	Name of receptor	Dry deposition rate ($\text{g m}^{-2} \text{s}^{-1}$) $V_d = w/C_o$
1.	Taj Mahal	0.22
2.	Agra Fort	0.34
3.	Itmal-ul-Daulah	0.30
4.	Nagar Maha Palika	0.23
5.	Sikandra	0.10

index of agreement, I , is expressed as

$$I = \frac{\sum_{i=1}^N (P_i - O_i)^2}{\sum_{i=1}^N (|P_i| + |O_i|)^2}$$

where $P_i = P_i - \bar{O}$ and $O_i = O_i - \bar{O}$.

Means are computed as

$$\bar{O} = \frac{1}{N} \sum_{i=1}^N O_i, \quad \bar{P} = \frac{1}{N} \sum_{i=1}^N P_i$$

where O_i refers to observed values and P_i refers to the corresponding predicted concentration at the same location during the same time period. N is the total number of observations.

The variable I specifies the degree to which the observed deviations about \bar{O} correspond, both in magnitude and sign, to the predicted deviations from \bar{O} (Rao *et al.*, 1985). The value of I for IITLT model is 0.6, whereas for CDM it is 0.54 (for a perfect model, $I = 1$).

4. CONCLUSIONS

The main objective of this study is to develop a simple operational model, capable of handling calm wind condition appropriately for planning and siting purposes.

In general, the output from the models (IITLT and CDM) are in reasonable agreement with the observed concentrations although there is a tendency towards overprediction.

The "calm wind conditions" occur very frequently in urban cities of India. With low wind speeds ($< 2.0 \text{ ms}^{-1}$), it is difficult to predict concentrations under these conditions from Gaussian models. In the present work attention has been given to calm wind conditions. A comparison of five different methods for treating calm winds with the IITLT model showed an improvement in the predicted concentrations.

Table 4 indicates that calm winds account for about 10% of the contribution to the concentration of pollutants. Since calm conditions are found to prevail about 60% of time, the importance of considering the contribution due to calm conditions is self-evident.

Other conclusions are outlined below.

(a) Intercomparison of five methods adopted to consider dispersion under calm conditions shows that method V incorporated in the IITLT model produces the best results (Table 4).

(b) The CDM takes 10 times more CPU time to run than the time taken by IITLT model.

(c) There is a substantial level of concentration of SO_2 and particulate matter in the Agra region. Possible sources are all coal users consisting of the two power plants, a number of small industries mainly foundries (approximately 250) and a railway shunting yard. As far as SPM is concerned because of use of

coal, contribution will be substantial. Even though the total amount of emission of SO_2 from these sources may be small, due to their proximity to the monuments, their contribution to the air quality of the zone near the monuments will be considerably high.

(d) In recent years, the two power plants have been shut down and a program of dieselization of the railway engines has begun. Control measures must take into account domestic use of coal and industrial uses of coal and fuel oil in planning a strategy for protecting the Taj Mahal from further deterioration.

(e) A study of Table 4 indicates that results from IITLT (without contribution due to calm conditions) overpredict the concentration which is understandable because Gaussian formulations are known to overpredict, incorporation of contribution due to calm winds in the IITLT model, will further increase the concentration, a trend discernible in Table 4.

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REFERENCES

- Adrian D. B and Zimmerman J. R. (1973) User's Guide for the Climatological Dispersion Model, National Environmental Research Centre, Office of Research and Development, U.S. EPA, Research Triangle Park, NC.
- Briggs G. A. (1969) Plume Rise, USAEC Critical Review Series, TID-25075, National Technical Information Service, Springfield, VA. 81 pp.
- Briggs G. A. (1971) Some recent analyses of plume rise observation. In *Proc. 2nd Intel. Clean Air Congress* (edited by Englund H. M. and Beery W. T.), pp. 1029–1032. Academic Press, New York.
- Briggs G. A. (1973) Diffusion estimation for small emission. NOAA Atmos. Turb. and Diff., Lab., Contribution File No. (Draft 79, Oak Ridge, TN. 59 pp.)
- Briggs G. A. (1975) Plume rise predictions. In *Lectures on Air Pollution and Environmental Impact Analysis* (edited by Haugen D. A.), pp. 59–111. American Meteorological Society, Boston, MA.
- Csanady G. T. (1973) *Turbulent Diffusion in the Environment*. Reidel, Dordrecht.
- Deardorff J. W. (1984) Upstream diffusion in the convective boundary layer with weak or zero mean wind. Conference volume, Fourth Joint Conference on Application of Air Pollution.
- Debberdt W. R., Ludwig F. L. and Johnson W. B. Jr (1973) Validation and application of an urban diffusion model for vehicular pollutants. *Atmospheric Environment* 7, 603–618.
- Draxler R. R. (1979) An improved Gaussian model for long term average air concentration estimates. *Atmospheric Environment* 14, 597–601.
- EPA (1973) User's Guide for the Climatological Dispersion Model. EPA-R-4-73-024.
- Fox D. G. (1981) Judging air quality model performance—a summary of the AMS workshop on dispersion model performance. *Bull. Am. met. Soc.* 62, 599–609.
- Gifford F. A. (1961) Uses of routine meteorological observation for estimating atmospheric dispersion. *Nucl. Safety* 2, 47–51.
- Handerson B. and Seller (1984) *Pollution of our Atmosphere*. Adam Hilger, Bristol.

- Hanna S. R. (1974) Urban diffusion problems. In *Lecture Notes on Environmental Impact Analysis*, pp. 209-224.
- Hanna S. R. et al. (1982) *Handbook on Atmospheric Diffusion*, Technical Information Centre, US Department of Energy.
- Industrial Pollution Control (1989) *Air and Water Revised*, Vol. I. Industrial Pollution Control Association of Japan, Tokyo, Japan.
- Ku J. Y., Rao S. T. and Rao K. S. (1987) Numerical simulation of air pollution in urban areas: model performance. *Atmospheric Environment* **21**, 213-232.
- McMohan T. A. and Danison P. J. (1979) Empirical atmospheric deposition parameters—A survey. *Atmospheric Environment* **13**, 571-585.
- Manju Kumari (1985) Diurnal variation of mean mixing depths in different months at Delhi. *Mausam* **36**, 71-74.
- Mathura Refinery Report (1978) Baseline air quality survey at Agra by NEERI, pp. v-2-v-33.
- Pasquill F. (1961) The estimation of the dispersion of wind borne material. *Met. Mag.* **90**, 33-49.
- Pasquill F. (1974) *Atmospheric Diffusion*, 2nd edition. Ellis Horwood, Chichester.
- Raghaven N., Pramila G. and Swati B. (1983) A Gaussian model for predicting SO₂ concentration in the city of Agra. *Atmospheric Environment* **17**, 2199-2203.
- Rao S. T., Sistla C., Pagnotti V., Petersen W. B., Irwin J. S. and Turner D. B. (1985) Evaluation of the performance of RAM with the regional air pollution study data base. *Atmospheric Environment* **19**, 229-245.
- Smith M. E. (1968) Recommendations for prediction of air borne effluents ASME IX, 85.
- Truppi L. E. (1968) Bias introduced by anemometer starting speeds in climatological wind rose summaries. *Mon. Wea. Rev.* **96**, 325-327.
- Turner D. B. (1969) Workshop of Atmospheric Dispersion Estimates, Washington, DC, HEW.
- U.P. Government Annual Statistics (1981) Coal data from supply office of Agra District. U.P. Government Publication.
- Willmott C. J. (1982) Some comments on the evaluation of model performance. *Bull. Am. met. Soc.* **63**, 1309-1313.