



Development and performance evaluation of statistical models correlating air pollutants and meteorological variables at Pantnagar, India

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ARTICLE INFO

Article history:

Received 11 April 2010

Received in revised form 16 October 2010

Accepted 2 December 2010

Keywords:

Air pollution

Multiple linear regression

Model performance

Model validation

Statistical analysis

ABSTRACT

Ambient air quality in respect of SO₂, NO₂ and total suspended particulate matter (TSPM) was monitored at Pantnagar, India from May, 2008 to April, 2009 and statistically analyzed with meteorological variables such as relative humidity (RH), wind speed (WS), precipitation (P) and mean air temperature (T). TSPM was found to be the major air pollutant causing significant deterioration of air quality with annual mean concentrations of 280 µg/m³. Further, weekly mean air pollutant concentrations were statistically analyzed through stepwise multiple linear regression analysis in respect of independent meteorological variables to develop suitable statistical models. Both NO₂ and TSPM concentrations were found to have been influenced by meteorological variables with coefficient of determination (R^2) of 82.21 and 92.84%, respectively. However, atmospheric SO₂ revealed only 22.87% of dependencies on meteorological variables. Partial correlation coefficients revealed that wind speed has the maximum influence (77.80 and 31.50%) on proposed equations for NO₂ and SO₂, closely followed by weekly mean temperature (73.60 and 24.30%). However, in case of TSPM, individual contribution of ambient temperature (94.40%) was found maximum, followed by relative humidity (86.50%). Model performances were evaluated through both quantitative data analysis techniques and statistical methods. Nearly 98 and 95% of potential error has been explained by the model developed for TSPM and NO₂, while in case of SO₂, it is found as only 61%. Therefore, performances of models (for TSPM and NO₂) to predict ambient weekly mean concentrations based on forecasted weather parameters were found to be excellent, however, performance of model developed for SO₂ was found only satisfactory.

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1. Introduction

In recent years, continuous industrial establishment, population expansion and increase in energy consumption have lead to the general deterioration of ambient air quality in most of the countries. Poor air quality has both acute as well as chronic health impacts (Nastos et al., 2010) and the severity of the impacts mostly depends upon two variable factors viz. ambient concentrations of the air pollutants and its exposure time. Further, concentrations of air pollutants are subject to

alter depending on the local topography, source emission and surrounding meteorological conditions. However, among these variables, meteorological parameters are chiefly responsible for causing variations in the ambient concentrations of air pollutants (Banerjee and Srivastava, 2009a). It is now well established that unfavorable meteorological factors like atmospheric stability, can greatly aggravate the impacts caused by certain air pollutants. Therefore, over the past few decades, air quality in the urban areas has been interpreted with the combination of various meteorological factors (Olcese and Toselli, 1997; Tasdemir et al., 2005; Ilten and Selici, 2008). Chao (1991) investigated the relationships between air pollutants concentrations with meteorology which has helped in formulating appropriate policies to combat air pollution in Shanghai,

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China. Escourrou (1990) explored the influence of different climatic scales viz. general, local and regional climatic pattern on air pollutants in Paris, France. Miyazaki and Yamaoka (1991) established relationship between mean atmospheric dust concentrations with some climatic variables in Osaka, Japan. Tirabassi et al. (1991) concluded the presence of significant correlation between wind speed and ground-level concentrations of air pollutants in the city of Ravenna, Italy. Ilten and Selici (2008) reported that ambient TSPM and SO₂ concentrations are highly correlated with meteorological variables in Balikesir, Turkey. Giakoumi et al. (2009) indicated a significant relation of suspended particles and nitrogen oxides concentrations with meteorological variables in Athens, Greece. Içağa and Sabah (2009) found considerable dependencies between air pollutants and humidity, temperature and inversion, but no relationship between wind velocity and precipitation in Afyon, Turkey. However, Cuhadaroglu and Demirci (1997), through multiple linear regression analyses reported the absence of any strong relationships between ground-level air pollutants with meteorological factors in urban areas of Trabzon, Turkey. Tasdemir et al. (2005) supported these findings by their experiment in Bursa, Turkey, where weak correlation was found to persist between meteorological parameters and air pollutant variables.

In urban areas, combustion of fossil fuels to generate electricity, in industrial processes, transportation and space heating is the predominant anthropogenic source of atmospheric air pollutants (CO, NO₂, SO₂, TSPM etc.). TSPM is the general term that is used to denote the solid particles and liquid droplets present in the atmosphere. These are diverse in physical and chemical properties depending on their source (stationary, mobile or natural), geography and meteorology of the particular area. Epidemiological studies emphasized that exposure to airborne particles with aerodynamic diameter <10 µm (Respirable Particulate Matter, RPM) induce negative health impacts (Nastos et al., 2010) and adverse meteorological factors may aggravate such kind of exposure. In addition to these, particulates are also responsible for causing reduced visibility and changes in the nutrient balance both through wet as well as dry deposition processes (US EPA, 1998). Atmospheric sulphur is one of the prominent anthropogenic pollutants, responsible for causing formation of sulphate aerosols, deposition of sulphate particles and generation of sulphuric acids droplets (H₂SO₄) under the apposite atmospheric conditions. Researches involving asthmatics indicated that significant proportion of population experience changes in pulmonary function and respiratory symptoms after very short periods of exposure to SO₂ (WHO, 2005). Sulphur-dioxide mostly generated through combustion of fossil fuels is therefore, closely associated with urban air pollution problems. Nitrogen oxides are the byproducts of fossil fuel combustion processes, therefore, emission from automobiles are significant contributors of atmospheric NO₂. Irritation in the respiratory tract is the predominant impact associated with NO₂ when it is converted to nitrates and nitric acid. However, protracted exposure may cause adverse impacts on the lung structure, metabolism and resistance against infections (Ali and Athar, 2008).

Elaborative knowledge on the effects of air pollutants on human health is a prerequisite for the development of effective air quality management policies. Geographical

analyses have previously been used in the epidemiological studies in order to establish relationships between human health consequences and air pollution exposure. However, variability of air pollutant concentrations in a location has different characteristics based on the prevalent meteorological conditions (Olcese and Toselli, 1997). Moreover, as meteorological factors differ significantly under varying geographical conditions, it is therefore, essential to study the impact of meteorology on the variation of ground-level pollutant concentrations. The present study was therefore, carried out at the Crop Research Centre (CRC) of G.B. Pant University of Agriculture and Technology, Pantnagar, India to statistically evaluate the relationship between the monitored ambient air quality with meteorological variables. Although, the experimental site is basically an agricultural research station, but the development of Integrated Industrial Estate-Pantnagar (IIE-Pantnagar) adjacent to this research centre resulted in gradual increase of regional pollution (Banerjee and Srivastava, 2009a, 2011). Moreover, the climatic and geographic features of the study area are also unique as it located in the Tarai region of Himalayas and accounts for varying climatic extremes throughout the year. In order to accomplish the objectives, the characteristics of the local topography and climatic features were studied and further, seasonal trends of prevailing ambient air quality were analyzed. Furthermore, concentrations of SO₂, NO₂ and TSPMs were investigated to establish statistical relationship with the meteorological variables viz. relative humidity (RH), wind speed (WS), precipitation (P) and weekly mean temperature (T). Conclusively, to evaluate the model performance and applicability of the proposed models in the study area, monitored and model predicted concentrations were statistically compared.

2. Characteristics of the study area

The climate of Pantnagar is characterized as humid subtropical, with high temperatures during summers (March to June), intense rainfall during monsoons (July to September) and severe cold in winters (December to February). Pantnagar is located with latitude 29°01'09"N and longitude 79°28'58"E, typically in the Tarai belt of Himalayas with an altitude of 243.8 m above the mean sea level. Annual average rainfall of the study area varies between 1300 and 1400 mm and 80% of which is received during the monsoonal season. In the year 2008, maximum and minimum recorded temperatures in this region are 41.2 °C and 1.7 °C, respectively (Banerjee and Srivastava, 2009a, 2009b).

3. Materials and methods

3.1. Sample collection and analysis

Hi-volume sampler (HVS, APM 410, Envirotech, Delhi) was used to monitor TSPM, SO₂ and NO₂ at CRC, Pantnagar. The TSPM was measured through GF/A Glass microfiber filter paper (Whatman, U.K.) of 8"×10" size. Filter paper was conditioned in a dessicator for 24-h and weighed on a balance (Precisa, Germany) with the sensitivity of 0.001 g, both before and after ambient air quality monitoring. The conditioned and weighed filter paper was placed in cloth-

lined envelope and used to avoid any possibilities of contamination and moisture adsorption. The TSPM monitoring was performed at an average flow rate of 1.2 m³/min. In order to maintain the specific flow rate, the manometer reading was taken 3–4 times in a day so that the flow rate variations were within 1.1–1.3 m³/min and finally an average flow rate was computed. Ambient air quality monitoring was done twice in a week continuously for 24-h. Adequate preventive measures were taken to avoid any sort of moisture adsorption to filter paper and concentrations of TSPM were calculated gravimetrically. Envirotech APM 411TE Thermo Electrically Cooled Gaseous Sampler (Envirotech, Delhi) attached with HVS was used to monitor the gaseous pollutants. For both SO₂ and NO₂, the monitoring was done at a constant flow rate of 1 lpm by bubbling ambient air through the liquid absorbing medium. Improved West and Gaeke method with Potassium-tetracholoromercurate (K-TCM) as absorbing medium (BIS: 5182 (Part 2), 2001) was used to determine ambient SO₂ concentrations. However, for determination of NO₂, modified Jacob and Hocheiser method (BIS: 5182 (Part 6), 2006) with solution of sodium hydroxide and sodium arsenite was used. Gaseous pollutants present in ambient air were absorbed in the respective absorbing medium and analyzed spectrophotometrically (Varian, U.S.) at 560 nm and 540 nm for SO₂ and NO₂, respectively (Banerjee and Srivastava, 2011).

The daily mean WS, RH, T and P for the study period of one year (May, 2008 to April, 2009) were collected from Meteorological Data Observatory at CRC, Pantnagar and computed monthly average values were illustrated in Table 1.

3.2. Statistical analysis

Relationship between the monitored air quality parameters and meteorological variables was determined through regression analysis using SPSS 15.0 statistical software programme. Gaseous pollutants and TSPM were considered as dependent variables, while meteorological parameters such as temperature, wind speed, relative humidity and precipitation were considered as independent. Pursuing the experiment, it was assumed that the dependent variables follow the normal distribution, homoscedasticity i.e., the data have the equal variance, and the difference between actual and theoretical values of dependent variables were independent (İçağa and Sabah, 2009). It was also assumed that the meteorological parameters used in the multiple variable analyses were independent of each other.

Linear regression analysis was performed to obtain the best probable prediction equation for the model chosen. This is mostly used to predict the coefficient of the linear equation involving one or more independent variables which are useful to estimate the value of the dependent variable

(Cuhadaroglu and Demirci, 1997; Ilten and Selici, 2008; İçağa and Sabah, 2009). A general regression equation having four independent variables (x_1, x_2, x_3, x_4) can be expressed as:

$$y = a + b_1x_1 + b_2x_2 + b_3x_3 + b_4x_4 \quad (1)$$

where a is the regression constant and b ($n = 1, 2, 3, 4$) is the regression coefficient. In order to minimize the error, the values of constant and coefficient are determined using the least-square method. The significance levels of the constant and the coefficient were statistically tested using t and F distribution. The analysis of the direction, strength and statistical meaning of the variables were then computed to find out the determination of coefficient (R^2). It is the fundamental measure of the goodness of fit of a linear model and defined as the proportion of total variability in the dependent variable that is accounted by the regression equation (Cuhadaroglu and Demirci, 1997; Ilten and Selici, 2008). It also defined as the 'explained variance' to the total variance. The explained variance is the part of the variance of the predicted data which is explained by the regression line (Juda, 1986). Therefore, R^2 can be expressed as:

$$R^2 = 1 - \frac{\sum (\hat{y}_i - \bar{y})^2}{\sum (y_i - \bar{y})^2} \quad (2)$$

where, \hat{y}_i is the value of y predicted by the regression line, y_i is the value of y observed, and \bar{y} is the mean of the y_i s. Values of the coefficient of determination equaling to 1 ($R^2 = 1$) signify that the fitted equation accounts for all the variability of the dependent variables. In contrast to this, $R^2 = 0$ indicates the absence of any linear relationship between the pollutants and meteorological variables. It is considered that a high value of R^2 assures a statistically significant regression equation and vice-versa (Norusis, 1990).

In the present study, stepwise multiple linear regression analysis was performed which is well established as an appropriate methodology to deal with the experiment undertaken (Cuhadaroglu and Demirci, 1997; Ilten and Selici, 2008; İçağa and Sabah, 2009). Stepwise regression is basically the combination of backward and forward procedure, where the first independent variable is selected against dependent variable in the same manner as in case of forward selection. The variable is examined for exclusion according to the removal criteria as in backward elimination. If the variable fails to meet the entry requirements (having lower values of R^2), it is removed until none of the remaining variables meets the removal criteria. If it passes the selection requirement, the second variable based on highest partial correlation is selected. The selection of independent variables terminates

Table 1
Monthly average value of meteorological parameters at CRC, Pantnagar.

Parameters	May	June	July	Aug.	Sep.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.
Wind speed (km/h)	9.25	6.25	5.66	5.25	3.96	2.69	2.15	2.46	3.87	5.78	5.61	7.68
Precipitation (mm)	1.15	13.91	18.74	26.67	13.85	0.82	0.11	0.00	0.00	0.00	0.09	0.60
Relative humidity (%)	47.13	73.08	82.42	82.35	76.48	67.97	65.95	71.27	74.77	71.05	58.50	44.45
Temperature (°C)	28.68	28.36	27.97	27.71	26.94	24.19	19.90	16.69	14.26	16.67	21.89	27.10

when no more variables meet the entry or removal criteria. In order to establish the correlations between the pollutant concentrations and pre-identified meteorological parameters by Eq. (1), the equation expressed as below (Eq. (3)) has also analyzed separately.

$$Y = f(X_1), Y = f(X_2), \dots, Y = f(X_2, X_3), \dots, Y = f(X_1, X_2, X_3, X_4, X_5) \quad (3)$$

During the process, the independent variables having lower values of R^2 have been eliminated and remaining variables were used to develop model equation. Finally, partial correlation was calculated to identify the individual contribution of the meteorological parameters on the developed models (İçaga and Sabah, 2009).

3.3. Model performance evaluation

The evaluation of air quality model performance focuses principally on the accuracy of the model prediction relative to the observed concentrations (Juda, 1986). In the present study, model equations were developed for three pre-identified air quality parameters based on the observed meteorological variables through statistical analysis. Therefore, a comprehensive analysis of the predicted model values in respect of observed concentrations is required to consider its applicability. The model performance was therefore, assessed by a comprehensive package of model accuracy prediction methods as suggested by Juda (1986) and Sivacoumar et al. (2001). There are several criteria for the evaluating the performance of a model and a brief description of the applied performance measures have been presented below.

a. Measures of model error:

i. Root mean square error (RMSE)

$$\text{Mean square error (MSE)} = N^{-1} \sum_i^n [O_i - P_i]^2 \quad (4)$$

$$\text{RMSE} = \sqrt{\text{MSE}} \quad (5)$$

ii. Systematic root mean square error (RMSE_s)

$$\text{MSE}_s = \text{RMSE}_s^2 = N^{-1} \sum_i^n [(a + bO_i) - O_i]^2 \quad (6)$$

where, a and b are the intercept and slope of regression equation $\hat{P} = a + bO$ [\hat{P} indicates the conditional mean value of P (predicted concentration) on a given O (observed concentration)], respectively and N is the number of data points.

Regression line has the form:

$$\hat{P} = a + bO \quad (7)$$

$$\text{where, } a = \bar{P} - b\bar{O} \quad (8)$$

$$\text{and } b = \left[\sum_{i=1}^N O_i P_i - N^{-1} \sum_{i=1}^N O_i \sum_{i=1}^N P_i \right] / \left[\sum_{i=1}^N O_i^2 - N^{-1} \left(\sum_{i=1}^N O_i \right)^2 \right] \quad (9)$$

The regression coefficients (both intercept and slope) were determined by fitting a first order polynomial

function to the concentration data using least-square method. The usefulness of the regression line depends to a large degree upon the extent to which actual predictions scatter away from it (Juda, 1986).

iii. Unsystematic root mean square error (RMSE_u)

$$\text{MSE}_u = \text{RMSE}_u^2 = N^{-1} \sum_i^n \left[P_i - (a + bO_i) \right]^2 \quad (10)$$

Systematic difference approach to zero signifies more accurate model performance, while the unsystematic difference should approach to RMSE as the system is conservative (Juda, 1986; Sivacoumar et al., 2001).

b. Noise to signal ratio (NSR):

Actual monitored values contain two basic components viz. signal, which one that carries the information of interest and noise, which other consists of random errors. The noise is superimposed on the signal component. The NSR is the ratio of the strength of the signal and noise and can be computed as:

$$\text{NSR} = \left[\frac{1}{N-1} \sum_{i=1}^N (O_i - \bar{O})^2 \right]^{1/2} / \bar{O} \quad (11)$$

where, N are the number of data points, O_i is the monitored concentrations of air pollutant, P_i is the model predicted concentration and \bar{O} is the average value of observed concentrations. The NSR should preferably be less than one for a good prediction model and as close to zero as possible.

c. Measures of correlation (r):

Monitored air quality parameter concentrations were further analyzed in respect of model predicted values through correlation analysis, which describes the quantitative association or agreement between predictions and observations. Coefficient of determination (R^2) between observed and predicted values was also computed. The formulae used for determination of correlation of coefficient (r) have the form:

$$r = \left[\frac{\sum_{i=1}^N (O_i - \bar{O})(P_i - \bar{P})}{\left[\sum_{i=1}^N (O_i - \bar{O})^2 \sum_{i=1}^N (P_i - \bar{P})^2 \right]^{1/2}} \right]^{1/2} \quad (12)$$

d. Index of agreement (d):

The measures of correlation to assess the performance of a model have some limitations. Often, it is found that small differences between monitored and model predicted concentrations results in negative values of correlation coefficient, which may be misleading while interpreting the model performance (Willmot and Wicks, 1980). The index of agreement (d) was originally proposed by Willmot and Wicks (1980) as an alternative to correlation coefficient (r). The index of agreement helps to determine the extent to which the observed variate is accurately estimated by the simulated variate and also suggests the percentage of potential for error that has been explained by the model (Sivacoumar and Thanasekaran, 1999). Its range varies from zero to one, while the later shows the

better model performance. The equation for computing index of agreement can be expressed as:

$$d = 1 - \left[\frac{\sum_{i=1}^N (O_i - P_i)^2}{\sum_{i=1}^N (|P_i - \bar{O}| + |O_i - \bar{O}|)^2} \right]. \quad (13)$$

4. Results and discussion

4.1. Observed monthly variations of air pollutant concentrations at CRC

Ambient air quality monitoring was performed on weekly basis and computed monthly variations of pollutants concentrations (SO_2 and NO_2) with reference to the pre-identified meteorological parameters (W, P, RH, T) are shown in Fig. 1. The figure illustrates that average temperature at Pantnagar usually persists within a higher range (19 to 29 °C), with a sharp decline during winter season (December to February). Further, a substantial portion of annual precipitation was received only during the monsoon season (July to September), whereas, higher values of relative humidity were measured during both monsoon as well as winter seasons. However, winter season was almost dry in respect of rainfall. Wind speed was relatively constant during the entire monitoring phase with slight variations in summer months. Monthly average ambient concentrations of TSPM have been plotted in Fig. 2 for May, 2008 to April, 2009. The 24-h and the annual prescribed standards for TSPM as recommended by the Central Pollution Control Board, India (CPCB, 2000) are 140 and 200 $\mu\text{g}/\text{m}^3$ for residential and commercial areas, respectively. It is well evident that average TSPM concentrations at CRC, Pantnagar were significantly above the annual standards with an approximate annual average concentration of 280 $\mu\text{g}/\text{m}^3$. As illustrated from the observed data, higher TSPM concentrations were experienced from the month of November and onwards, which were particularly intensive in winters (December to February). The probable reason behind these findings may be the persistence of stable atmospheric conditions during the winter season which restricts the dispersion of air pollutants (Banerjee and Srivastava, 2011). The average concentrations of gaseous pollutants (SO_2 and NO_2) for the monitoring period of one

year (May, 2008 to April, 2009) are presented in Fig. 3 (a and b). Annual average concentrations for SO_2 and NO_2 were approximately 8.7 and 10.6 $\mu\text{g}/\text{m}^3$, respectively, which were well below the NAAQS (the 24-h and annual standards are 80 and 50 $\mu\text{g}/\text{m}^3$ and 80 and 40 $\mu\text{g}/\text{m}^3$ for SO_2 and NO_2 , respectively) (CPCB, 2010). Observed concentrations for both these gaseous pollutants reveal a general trend of winter time maximum and summer time minimum. A plausible explanation for such observations may be found by scrutinizing the existing meteorological conditions at the monitoring location. The most considerable factors related to the dispersion of air pollutants are speed of wind and the extent to which the emission can rise into the atmosphere (mixing height). The product of maximum mixing height and the average wind speed within the mixing height is recognized as atmospheric dispersive capability or ventilation coefficient. The ventilation coefficient usually remains at its maximum during the summers and resides at its minimum during the winter seasons. Moreover, the general meteorology of the region during the winter is dominated by high pressure causing increased atmospheric stability, which in turn allows for less general circulation and thus more stagnant air (Aneja et al., 2001; Banerjee and Srivastava, 2011). Therefore, during winters, dispersion of atmospheric pollutants remains typically at its minimum and consequently elevated levels of pollutant concentrations are achieved. Conversely, during the summers, the average mixing height remains at its highest naturally, resulting mixing through a greater volume of air and hence resulting in lower pollutant concentrations. Additionally, reduction of precipitation during winters reduces the potential for wet deposition and associated cleaning mechanisms.

4.2. Statistical relationship between meteorological variables and air quality parameters

The correlation between individual air pollutants and meteorological variables (W, P, RH and T) from May, 2008 to April, 2009 were computed and shown in Table 2. Further, the relationships between weekly mean SO_2 , NO_2 and TSPM with meteorological parameters were analyzed by stepwise multiple regression analysis through application of SPSS 15.0 statistical package. The correlation coefficients (r) between SO_2 , NO_2 and TSPM with meteorological parameters describe

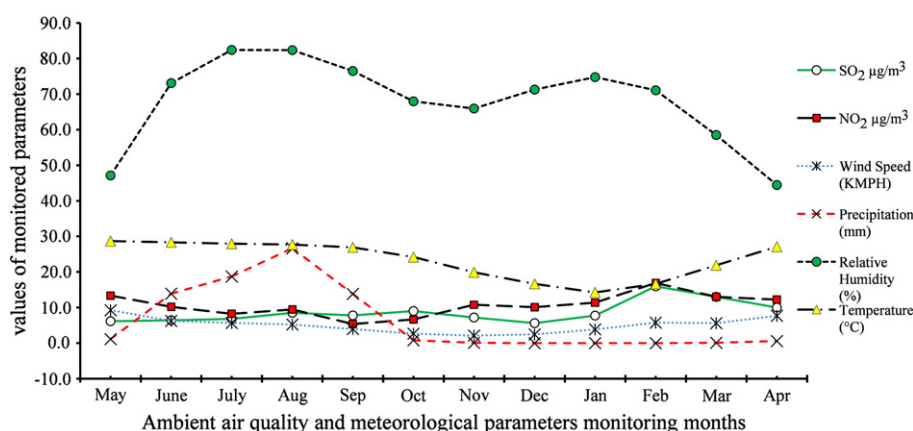


Fig. 1. Monthly variations in the concentrations of gaseous pollutants with reference to meteorological parameters.

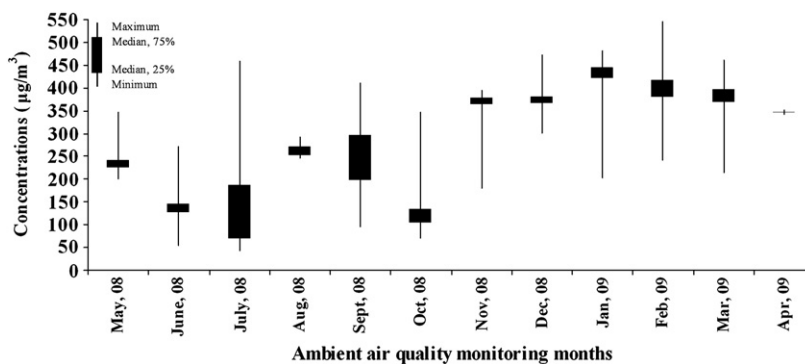


Fig. 2. Monthly variation in TSPM concentrations ($\mu\text{g}/\text{m}^3$) at CRC, Pantnagar.

the presence of single statistically significant negative correlation ($p < 0.01$) between TSPM and temperature, and do not exhibit further significant relationship with any other meteorological parameters. It is obvious that the pollutant concentrations should decrease effectively with increasing temperature as such can increase the possibility of enhanced wind circulation and simultaneously dilution of pollutants. In the present study, correlation between concentrations of gaseous pollutants and wind speed revealed contrasting results with the findings of majority of earlier reports (Turalioğlu et al., 2005; Ilten and Selici, 2008) but resembles with the findings of İçağa and Sabah (2009) (Table 3). Concentrations of SO_2 and NO_2 exhibit a positive correlation

(0.138 and 0.470, respectively) with wind speed, which might well be influenced by the presence of IIE-Pantnagar in the vicinity of the monitoring location. The selected monitoring location at CRC is located at 8.1 km in the downwind direction of IIE-Pantnagar, which is responsible for gradual deterioration of regional ambient air quality (Banerjee and Srivastava, 2009a, 2011). Pantnagar is itself a semi-urban area with no major air pollution source and therefore, the recent establishment of industrial estate and other adjacent commercial activities at the IIE-Pantnagar may be held accountable for deterioration of ambient air quality.

It can be seen from Fig. 1 that the monthly average values of relative humidity remain at their maximum during both

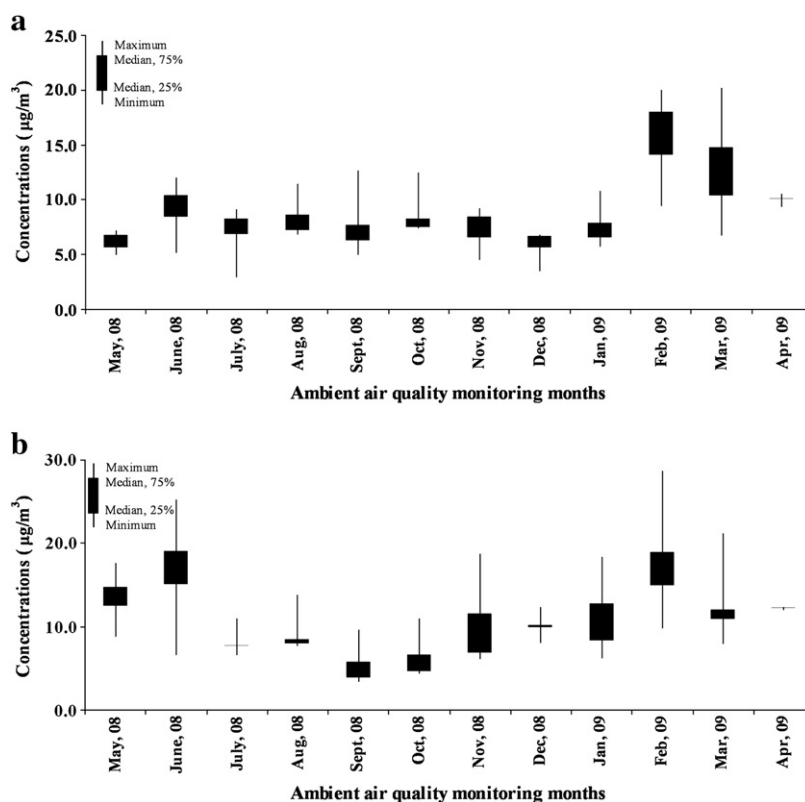


Fig. 3. a. Monthly variation in SO_2 concentrations ($\mu\text{g}/\text{m}^3$) at CRC, Pantnagar. b. Monthly variation in NO_2 concentrations ($\mu\text{g}/\text{m}^3$) at CRC, Pantnagar.

Table 2Correlation coefficients (*r*) between air quality parameters and meteorological variables.

Dependent variables	Wind speed	Precipitation	Relative humidity	Temperature
SO ₂	+0.138	−0.279	−0.145	−0.307
NO ₂	+0.470	−0.482	−0.459	−0.383
TSPM	−0.109	−0.561	−0.249	−0.762**

Note: **Correlation is significant at the 0.01 level (two-tailed).

monsoon and winter seasons, and therefore, a strong relation between air pollutants and humidity ratio was not expected. A weak negative correlation ($p < 0.05$) was observed for relative humidity with SO₂, NO₂ and TSPM, and the observations were in line with the findings of other researchers (Cuhadaroglu and Demirci, 1997; Ilten and Selici, 2008). The findings of other researchers related to correlations between air pollutants and meteorological variables are given in Table 3. It can be well demonstrated that correlations of different air pollutants and meteorological variables somewhat differ in different related researches and may be subject to variations depending upon the location and its unique meteorological characteristics. Table 2 describes that the pattern of correlation between SO₂ and NO₂ with meteorological variables somewhat resembles each other with slight difference from that of TSPM. Therefore, only the annual concentrations of SO₂ and TSPM as a function of meteorological parameters are graphed in Figs. 4 (a to d) and 5 (a to d), respectively.

The findings of stepwise multiple regression analysis between meteorological parameters and the air pollutants are illustrated in Table 4. In the experiment, initially, relationship between the individual air pollutant and all meteorological variables was determined stepwise and further, individual air pollutant was compared with each meteorological parameter in order to develop the most efficient statistical model. Mathematical functions illustrating the dependencies of TSPM, SO₂ and NO₂ with meteorological variables were compared for their statistical significance both at 5% and 1% levels. The most recommended equations were chosen based on the maximum values of determination coefficient (R^2) because all the investigated equations were significant at the 1% error level ($p < 0.01$). Therefore, the model equation for NO₂ concentration prediction including

the transformation of the dependent and the independent variables was formulated as:

$$\text{NO}_2 = 24.702 + 1.222(\text{WS}) + 0.102(\text{P}) - 0.089(\text{RH}) - 0.635(\text{T}) \quad (14)$$

$$r = 0.907 \text{ and } R^2 = 82.21(\%).$$

According to Eq. (14), the average temperature and relative humidity has negative influence on the ambient concentrations of NO₂ in contrast to that of wind speed and precipitation. The contribution of meteorological factors to the ambient concentrations of NO₂ at CRC, Pantnagar was found to be 82.21% and the remaining can be considered as indeterminate. The individual contribution of independent variable to the proposed equation was determined using the partial correlation coefficients (Table 5). For the anticipated equation, the variables WS, P, RH and T contribute 77.8, 21.3, 26.4 and 73.6% to the ambient NO₂ concentrations. The dominant source of ambient NO₂ in the study region is horizontal dispersion of gaseous pollutants from IIE-Pantnagar through wind and therefore, computed partial correlation coefficients were according to the expected one. Although the developed equation (Eq. (14)) reveals that precipitation exerts a positive influence on NO₂ concentrations, however, correlation coefficients between NO₂ and precipitation (Table 2) demonstrate presence of negative correlation (−0.482) between these two variables. The observed correlation was according to the expected one as the possible impact of precipitation on ambient NO₂ concentration should be negative as precipitation acts as wet scavenger of air pollutants. Further, temperature has obvious effects on pollutant concentrations by causing variations in wind circulation and simultaneously diluting of concentrations of pollutants (Banerjee and Srivastava, 2011).

Partial correlation coefficients among all the independent variables signify that contribution of wind speed is the most significant factor causing variation of ambient NO₂ concentrations which establish the fact that dispersion of NO₂ from adjacent IIE-Pantnagar towards CRC, Pantnagar is mainly causing enhanced level of NO₂ concentration. Relation between meteorological variables and atmospheric NO₂ has originated to be strong and thus the developed equation can be useful for quantitative prediction of NO₂ concentration based on forecasted weather at CRC, Pantnagar.

Table 3Correlation coefficients (*r*) as illustrated by other researchers.

Air pollutants	Wind speed	Precipitation	Relative humidity	Temperature	References
SO ₂	+0.005	−0.158	+0.418	−0.559	Içağa and Sabah (2009)
TSPM	+0.047	−0.141	+0.470	−0.622	Içağa and Sabah (2009)
SO ₂	−0.2615	–	+0.09	−0.51645	Ilten and Selici (2008)
TSPM	−0.43126	–	+0.28046	−0.30342	Ilten and Selici (2008)
SO ₂	−0.493	−0.137	+0.028	−0.755	Turalioğlu et al. (2005)
TSPM	−0.640	−0.075	+0.130	−0.795	Turalioğlu et al. (2005)
SO ₂	0.88	–	–	−0.42	Gupta et al. (2003)
SO ₂	(−0.13) to (−0.42)	–	0.03–0.37	(−0.39) to (−0.68)	Bridgman et al. (2002)
SO ₂	−0.46	–	−0.32	–	Kartal and Özer (1998)
Smoke	−0.55	–	−0.52	–	Kartal and Özer (1998)
RPM	(−0.15) to (−0.35)	–	0.08 to (−0.058)	(−0.05) to (−0.65)	Monn et al. (1995)

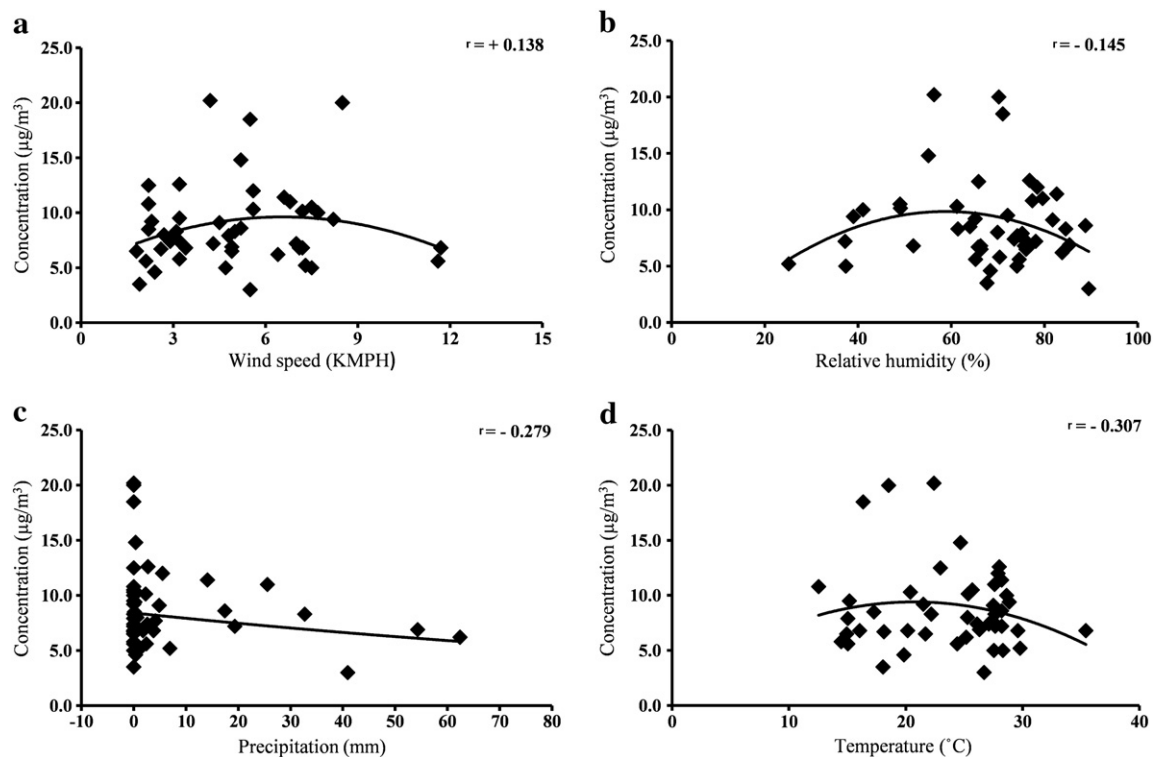


Fig. 4. SO_2 concentration versus: a. wind speed; b. precipitation c. relative humidity; d. temperature.

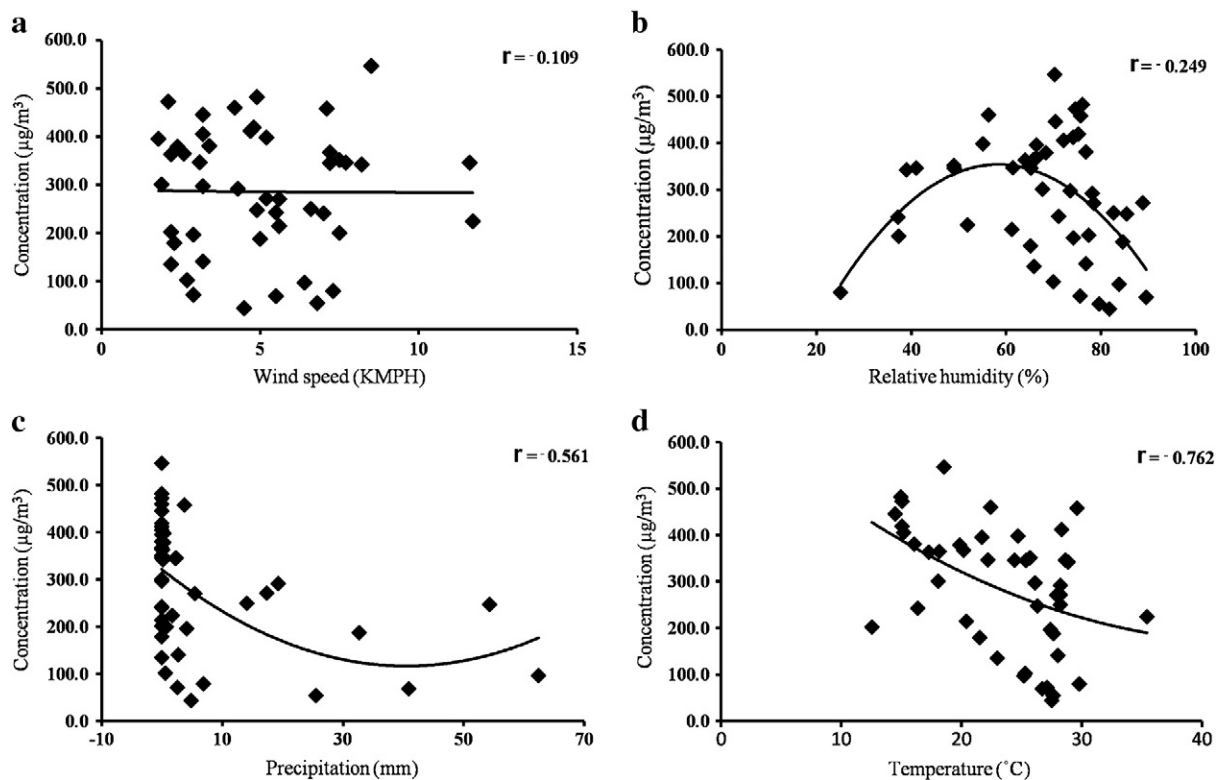


Fig. 5. TSPM concentration versus: a. wind speed; b. precipitation c. relative humidity; d. temperature.

Table 4

Results of regression analysis and equations between air pollutants and meteorological parameters.

State	R ² (%)	p value	Equation
SO ₂ with WS	1.904	0.01	$-0.273(WS)^2 + 3.158(WS) + 0.883$
SO ₂ with P	7.784	0.01	$0.012(P)^2 - 0.371(P) + 9.542$
SO ₂ with RH	2.103	0.01	$8.743e^{-0.00(RH)}$
SO ₂ with T	9.425	0.01	$-0.060(T)^2 + 2.477(T) - 14.69$
NO ₂ with WS	22.090	0.01	$0.025(WS)^2 + 0.404(WS) + 7.849$
NO ₂ with P	23.232	0.01	$0.016(P)^2 - 0.532(P) + 12.00$
NO ₂ with RH	21.068	0.01	$-0.002(RH)^2 + 0.243 + 7.618$
NO ₂ with T	14.669	0.01	$0.023(T)^2 - 1.246(T) + 26.50$
TSPM with WS	1.188	0.01	$-24.21\ln(WS) + 323.3$
TSPM with P	31.472	0.01	$0.744(P)^2 - 22.22(P) + 336.8$
TSPM with RH	6.200	0.01	$-0.138(RH)^2 - 15.62(RH) - 117.3$
TSPM with T	58.064	0.01	$-0.061(T)^2 - 10.65(T) + 570.1$

Note: WS: wind speed (km/h); T: temperature (°C); RH: relative humidity (%); P: precipitation (mm).

However, in contrast to NO₂, weekly average concentrations of SO₂ revealed a statistically insignificant relation with that of precipitation and relative humidity and consequently both has been eliminated from the equation. Therefore, the regression equation for SO₂ can be expressed as:

$$SO_2 = 12.924 + 0.622(WS) - 0.315(T) \quad (15)$$

$$r = 0.478 \text{ and } R^2 = 22.87\%.$$

The proposed equation illustrates that ambient SO₂ is only dependent on wind speed as well as average temperature, and both are cumulatively responsible for only 22.87% of SO₂ concentrations. The remaining ambient concentrations of SO₂ were independent of meteorological factors. Previous case studies on source contribution of SO₂ in the study region established that contribution of IIE-Pantnagar was insignificant in contrast to local domestic sources, and this may be the probable reason behind less dependence of SO₂ concentrations on meteorological factors (Banerjee and Srivastava, 2009a). The computed partial correlation coefficients also suggests that contribution of wind speed is the maximum (31.3%) for causing variations in ambient concentrations of SO₂, closely followed by temperature (24.3%). Except these two, remaining meteorological variables are not found to be significant contributors.

The existence of IIE-Pantnagar near to CRC is mostly considered to be decisive factor causing enhanced level of concentrations of gaseous pollutants. However, except NO₂, contribution of IIE-Pantnagar for regional burden of SO₂ concentration might not be found statistically significant. Preliminary research on dispersion of SO₂ surrounding IIE-

Pantnagar revealed that industrial source impact on SO₂ concentrations tends to get gradually reduced from 2007 to 2009 (Banerjee and Srivastava, 2009a, 2011). However, in order to establish domestic sources impact on ambient SO₂ concentrations, elaborative research will be carried out in the future.

From the monitored annual data, the regression equation obtained for TSPM in respect of meteorological variables is:

$$TSPM = 1647.78 + 13.911(P) - 10.467(RH) - 31.601(T) \quad (16)$$

$$r = 0.964 \text{ and } R^2 = 92.84\%.$$

According to Eq. (16) ambient concentrations of TSPM depend on meteorological variables by 92.84%. All the pre-identified independent variables except wind speed are found to have statistically significant relationships ($p > 0.01$) with the ambient concentrations of TSPM. Computed correlation coefficients between precipitation and relative humidity with TSPM were -0.561 and -0.249 , which establish the significance that both variables act as a wet scavenger of atmospheric aerosols. In order to establish the individual contribution of respective meteorological variables towards TSPM concentrations, the partial correlation coefficients were also compared. According to these coefficients, the variable temperature contributes 94.4% to the TSPM dependent variable. Individual contributions of precipitation and relative humidity were also significant causing 84.5 and 96.5% variations in ambient TSPM concentrations. In contrast to the other two developed models, implication of wind speed towards dependent variable TSPM was minimum (2.8%).

Comparing individual coefficients of determination (R^2) for each proposed equation, it can be concluded that in both the case of NO₂ (82.21%) as well as TSPM (92.84%), the developed equation may be considered as appropriate to compute weekly mean concentrations depending on the forecasted weather or measured meteorological observations. However, the proposed equation for computing SO₂ may not be as efficient due to lower levels of interaction (22.87%) between the ambient concentrations and meteorological variables. Domestic source contribution on atmospheric SO₂ concentration in respect of NO₂ and TSPM may be the possible reason behind such observations. All the proposed equations were further used to compute the expected values of the air quality parameters and deviations from the observed concentrations and have been plotted in Fig. 6 (a to c) for NO₂, SO₂ and TSPM, respectively.

4.3. Model performances evaluation

The results of the model simulation are summarized in Table 6. The evaluation of the performance of an air quality model generally focuses on assessing the accuracy of the model's predictions relative to observations. However, it is important to note that this approach represents only a part of the overall model evaluation problem. Therefore, besides only comparing the average values of the predicted and observed concentrations, model performance was evaluated on the basis of outcome of the seven statistical indicators and further compared with ideal value of a perfect model as suggested by Juda (1986).

Table 5

Partial correlation coefficients between air quality parameters and meteorological variables.

Dependent variables	Wind speed	Precipitation	Relative humidity	Temperature
SO ₂	0.313	-0.019	0.026	-0.243
NO ₂	0.778	0.213	-0.264	-0.736
TSPM	-0.028	0.845	-0.865	-0.944

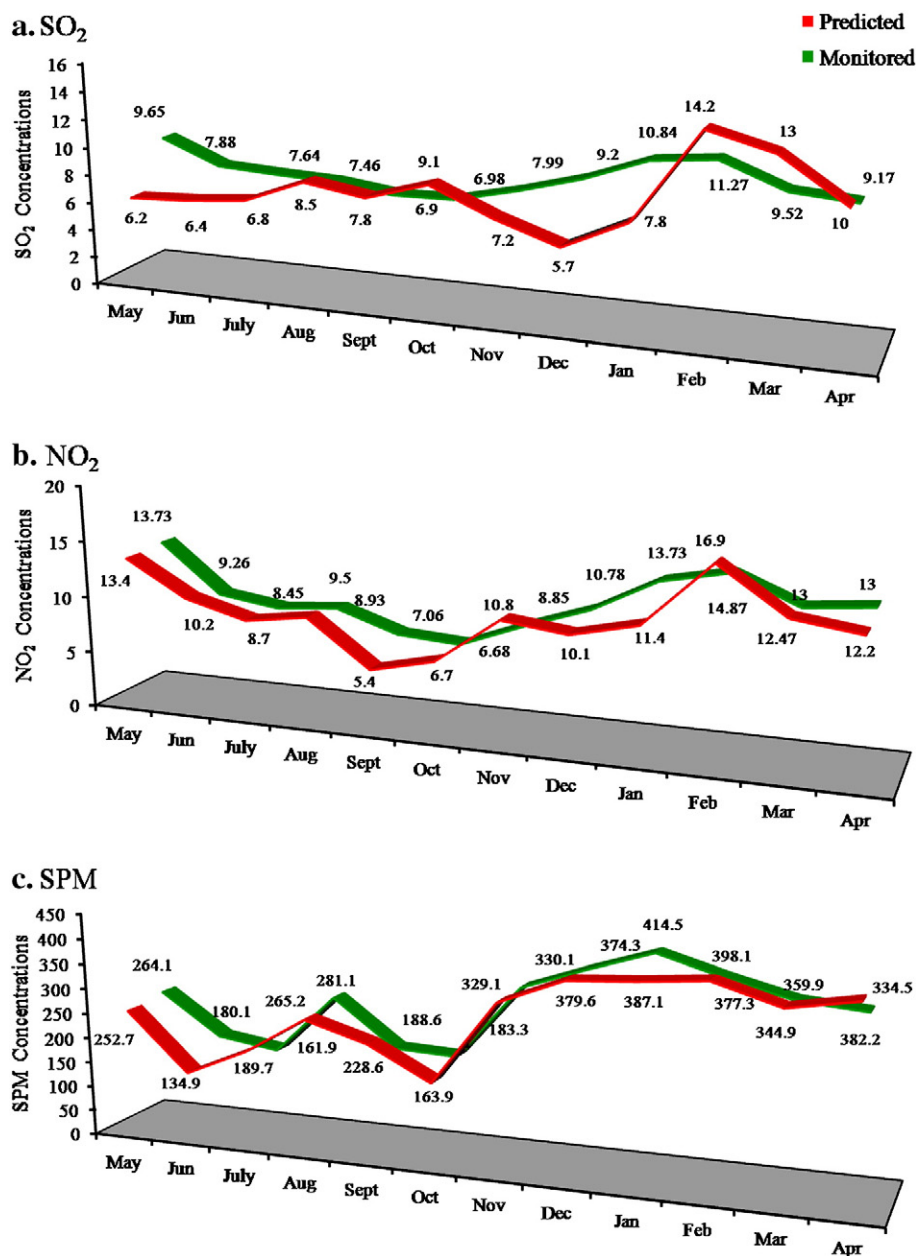


Fig. 6. Comparative analysis of model performance with observed concentrations: a. NO₂; b. SO₂ c. TSPM.

The difference measures, such as RMSE_u or RMSE_s, generally agree with the univariate summary measurements with regards to capabilities of the model to predict concentrations (Sivacoumar and Thanasekaran, 1999). The unsystematic errors (RMSE_u) of all three developed models approached towards their individual mean errors (RMSE), whereas systematic errors (RMSE_s) were relatively small except the model formulated for TSPM. Therefore, model performances in respect of computing of error were found satisfactory. Model predicted concentrations also expressed significant correlation with observed concentrations for both

SO₂ and TSPM (except that of NO₂), for which the correlation of coefficient varied from 0.436 to 0.955. Computed NSR for all developed models also explained satisfactory outcome suggesting the presence of less random errors in predicting the ambient concentrations of air pollutants. The regression coefficients (both intercept and slope) were determined by fitting a first order polynomial function. In case of both NO₂ and TSPM prediction models, the regression coefficients *b* (slope) were 0.94 and 1.02, respectively, indicating their satisfactory prediction of air quality. However, the model formulated for SO₂ expressed a poor regression coefficient *b*

Table 6

Model performance evaluation.

Parameter	Units	Ideal value (for perfect model)	Model for SO ₂	Model for NO ₂	Model for TSPM
RMSE	µg/m ³	–	2.32	1.25	27.21
RMSE _s	µg/m ³	0.0	0.32	0.17	3.47
RMSE _u	µg/m ³	RMSE	2.30	1.26	26.99
NSR	–	0.0	0.28	0.12	0.09
Coefficient of determination (R ²)	(%)	100	81.72	19.00	91.20
Correlation coefficient (r)	–	1.0	0.904	0.436	0.955
Regression coefficient 'a' (intercept)	µg/m ³	0.0	1.59	0.69	–2.7
Regression coefficient 'b' (slope)	(/)	1.0	0.80	0.94	1.02
Index of agreement (d)	%	100	60.80	94.83	97.57

with 0.80. Regression coefficient *a* (intercept) for all the developed models differed somewhat from that of perfect model and lied within the range of –2.7 to 1.59.

The performance of proposed air quality models was finally evaluated by computation of index of agreement (*d*). It provides the degree to which the model predictions are error free, by assessing the accuracy of the predicted variate in respect of observed concentrations. The analysis revealed that 95% of the potential for error has been explained by the model developed for computing NO₂ concentrations, while in case of TSPM model, the value goes further to 98%. Thus, for both the models (NO₂ and TSPM), the performance can be considered as excellent. However, index of agreement for the model used for computing SO₂ was 61%, signifying performance as only satisfactory. The principle explanation for such kind of performance by model used for SO₂ may be explained by the coefficient of determination (*R*²) between ambient SO₂ concentrations and meteorological variables. As seen from Eq. (15), only 23% of SO₂ depends on meteorological variables, while the remaining portion (77%) is indeterminate. Thus the level of relation between ambient SO₂ and meteorological parameters can only be considered as weak. However, significant levels of interaction were achieved for the cases of NO₂ and TSPM with meteorological parameters, where levels of interaction were 82 and 93%, respectively (Eqs. (14) and (16)). Thus, statistical analyses indicate that the accuracies of the developed models for SO₂, NO₂ and TSPM were about 61, 95 and 98%, and the relevant analysis of RMSE illustrates the presence of less error in the model prediction.

Atmospheric concentrations of all pre-identified pollutants are not solely influenced by meteorological variables, but, source contributions, generation of secondary pollutants and long-range transport. Therefore, atmospheric concentrations of these pollutants cannot be explained only in terms of persisting meteorology. However, the proposed equations can be very useful for broad reconnaissance as a first step to identify the locations and areas of concern through simple analysis of routinely forecasted meteorological variables.

5. Conclusions

A comprehensive study on dependencies of air quality parameters on meteorological variables were investigated at CRC, Pantnagar, India during May, 2008 to April, 2009. Among all the monitored air quality parameters, TSPM was originated to be the most critical pollutant and all the pollutants

exhibit the general trend of winter time maximum and summer time minimum concentrations.

In addition to these, the statistical relationship between weekly mean TSPM, NO₂ and SO₂ concentrations with meteorological factors, such as W, P, RH and T was statistically analyzed using the stepwise multiple linear regression analysis. Both NO₂ and TSPM concentrations were found to have well influenced by meteorological variables with coefficients of determination 82.21 and 92.84%, respectively. However, atmospheric SO₂ revealed only 22.87% of dependencies. Ambient concentrations of gaseous pollutants exhibit a positive correlation with wind speed, well influenced by the existence of IIE-Pantnagar in upwind direction in respect of CRC, Pantnagar. Outcome of partial correlation coefficient revealed that among all other meteorological factors, wind speed has the greatest influence on proposed equation, closely followed by ambient temperature. The plausible explanation of such observations may be variations in source contributions from that of IIE-Pantnagar and domestic sources.

Conclusively, the proposed equations for air quality parameters were tested to evaluate its performance. Comparison between model outputs and monthly average pollutant concentrations were made using both quantitative data analysis methods and statistical procedures. The analysis revealed that 95% of potential for error has been explained by equation developed for NO₂, while in the case of equation for TSPM, it reaches to 98%. However, model used for computing SO₂ concentration revealed only satisfactory performance with 61% of index of agreement. The probable reason behind such kind of observation may be explained by the poor interaction (*R*² = 23%) between meteorological factors and ambient SO₂ concentrations. Accuracies of developed equations for TSPM, NO₂ and SO₂ were found to be 98, 95 and 61% and therefore, concluded that the proposed equations may well be suitable for predicting air pollutants concentrations based on forecasted weather.

Policy and ethics

The work described in this article do not poses any experiments involving humans.

Conflict of interest

The authors hereby clearly mention that there are no conflicts of interests including any financial, personal or

other relationships with other people or organizations within three years of beginning the submitted work that could inappropriately influence, or be perceived to influence, their work.

Contributors

Dr. T. Banerjee: He is the first and corresponding author of the article. The experiment described here is the outcome of his PhD research work carried out in Dept. of Environmental Science, GBPUAT, Pantnagar, India. His main responsibility was to design, execute and experiment the work in the field and also to prepare the draft manuscript.

Dr. S.B. Singh: He is the second author of the article and also responsible to carry out the statistical analysis for the development of the models.

Dr. R.K. Srivastava: He is the third author of the article and responsible to design and execute the entire experiment. Under his guidance, the first author carried out the entire experiment in the field.

All authors have approved the final article and provided their consent to submit the article to **Atmospheric Research**.

Role of funding source

This study has been undertaken as a part of the project on “Environmental Impact Assessment Study of SIDCUL Integrated Industrial Estate Pantnagar”, funded by Uttarakhand State Council for Science & Technology (UCOST), Dehra Dun, India. However, UCOST's sole responsibility was to support the project financially and the remaining accountability lies on Dr. R.K. Srivastava as the principal investigator of the project.

Acknowledgements

This study has been undertaken as a part of the project on “Environmental Impact Assessment Study of SIDCUL Integrated Industrial Estate Pantnagar”, funded by Uttarakhand State Council for Science & Technology, Dehra Dun. Authors are thankful to Head, Department of Environmental Sciences; Dean, C.B.S.H & Director Experiment Station, G.B.P.U.A.T., Pantnagar; Dr. PC Joshi, Resident Engineer, IIE-Pantnagar; SK Biswas and Dr. Kamal Pandey for their technical assistance during this study.

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