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Estimation of Ambient Air Quality in Delhi

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1. Introduction

For decades, it has been well known that air pollution causes adverse health effects on humans (Thurston et al., 1989, Shandilya and Khare, 2012). Since the late 1980s, epidemiological studies were published relating the mortality and morbidity to the ambient levels of fine particles without showing the expected results (Borja-Aburto et al., 1997). Most emphasis was then given to the particles with an aerodynamic diameter smaller than 10μm (PM_{10}) particularly to the class of the fine particulates. A special study carried out by United Nations Sources (UNECE, 1979) shows that the frequency of finding a particle of diameter range 2-4μm is 27.5%, which is higher in an overall range of 1-12μm, so the danger of the fine particulates of diameter ranges 2-4μm is higher than the danger by any particle which may lie in the range of 4-12μm. Therefore, there is a rising awareness about further fine fraction i.e., particles with an aerodynamic diameter smaller than 2.5μm ($PM_{2.5}$). Aerosols, which originate directly from the sources, are termed as "Primary aerosols". There may be the conversion of gaseous matter into the particulate due to many Physico-Chemical reactions. Such types of particulates are known as "Secondary Aerosols" (Perkins, 1974, Shandilya et al., 2009).

Delhi, the Capital and third largest city of India, ranks third in population among other Indian cities, estimated among 8.5 million (Crowther et al., 1990). Rapid urbanization and the unprecedented industrial and economic development during the last three decades have increased the vehicular population of Delhi by several folds. Consequently, harmful emission such as Pb, PAH, SO_2, NOx, particulate matter and carbon monoxide have also registered a sharp increase in these intervening years. As per the WHO report, Delhi is the fourth most populated city in the world. A 1997 study by the Center for Science and Environment (CSE) revealed that at least one person died prematurely every hour in Delhi in 1995 strictly because of Suspended Particulate Matter (SPM). A 1998 study by All India Institute of Medical Sciences (AIIMS), New Delhi, showed how emergency visits and deaths due to respiratory and heart problems are the highest when particulate levels peak during the winter (Down to Earth, 2000). It is estimated that in Delhi, the incidence of chronic

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bronchitis is 6-14 times more than the average elsewhere in India because of the thermal power plants and the vehicular pollution (Rao & Rao, 1989).

The objective of this study was to monitor NOx, SO_2, PM_{10}, $PM_{2.5}$, and Non Respirable Suspended Particulate (NRSP) levels in South Delhi at different land use patterns; to compare it with the levels found in the crowded residential site of East Delhi; and to compare the observed SPM,NOx, and SO_2 levels with the prescribed CPCB limits (except $PM_{2.5}$ compared with USEPA primary standards), which serves as the guidelines for the environmental planning of the region. Another objective was to find out the spatial and diurnal variation of NRSP, the Organic Matter level in NRSP, and the 12 hour variation of NOx and SO_2. The final objective was the determination of Air Pollution Index at different places.

2. Materials and methods

A background to the Study Area is outlined in detail in Shandilya et al. (2007). In order to assess ambient pollution level, particulate samples were taken with the help of respirable dust sampler and fine particulate sampler. For the high volume Measurement of RSP, air was drawn through a 20.3X25.4cm Whatman GF/A glass fiber filter at a flow rate which was kept around $1\text{-}1.1m^3/\text{min}$. Detailed Procedures of Equipment (APM 460DX) are specified in operating manuals, available from equipment manufacturers (Envirotech Instruments Private Limited). The gases were determined with APM 411. The entire weighing procedure was done under controlled conditions (room temperature) in a balance room at Envirotech Instruments Pvt. Ltd., New Delhi using a Dhona-100DS microbalance. For $PM_{2.5}$ monitoring, ambient air enters APM550 system through an omni directional inlet designed to provide a clean aerodynamic cut-point for particles >10μm.

2.1 Sampling sites and details

The air quality monitoring in Delhi for present investigation was carried out from October 19, 2000 to November 29, 2000, during which PM_{10}, $PM_{2.5}$, and non-respirable suspended particulate concentrations were measured at four monitoring sites. In order to obtain a clear picture of a source-based pollution, it was required that the monitoring must be carried out at different places of different characteristics in New Delhi. It was important to consider residential, industrial, and commercial areas for carrying out such source differentiation. Based on the above considerations, four sites were chosen for setting up the sampling sites, namely Envirotech Instruments Pvt. Ltd, Okhla, II Floor Roof (Industrial Area); Indian Institute of Technology, Delhi, V Block IV Floor Roof (Residential Area); Vasant Vihar Bus Depot II Floor Roof (Commercial Area); Shakarpur a Residence, III Floor Roof (Residential Area). More details and figures of sampling locations can be obtained from Shandilya et al., (2007, 2012).

Out of the four sites chosen, two were in residential areas, one each in industrial and commercial areas. The criterion that was taken into account was the presence of local sources. The Shakarpur site was of particular importance, since the results obtained were used to compare the SPM level in South Delhi. Simultaneously, when we analyzed the respirable suspended particulates in respect of heavy metals, it was interesting to know the concentration near prominent sources like the Indraprastha power station. The site was located just on the bank of the Yamuna, when on the other bank was the Indraprastha power station. But the site

location was mainly a residential area. Except for Shakarpur, where the Indraprastha power station is situated nearby, the sources of the local emissions were considered.

SPM was collected on a filter paper weighed before and after exposure. It was dried in an oven at 80ºC for 45 minutes before each weighing. The size classification of PM was achieved through a cyclone installed in a Hi-Vol sampler, which separates the respirable PM_{10} and non-respirable fraction in the case of the Hi-Vol sampler. The particles sized <10µm were collected on the filter, and >10µm were collected in a separate sampling cup, also known as a dust collector. Flow rates were monitored before and after every sampling period with manometer, which was factory calibrated with an accuracy of 1.3% full scale. The flow rate checks were made before and after each sampling event to ensure that filter holders were not leaking and to determine that fitter clogging had not occurred.

These gases were monitored with the help of APM411 assembly attached with APM460DX (Envirotech Instruments Pvt. Ltd.) using the impingers. The modified Jacob-Hochheiser method (or arsenate method) have determined nitrogen dioxide (IS-5182, 1975). For the 24 hour field sampling, it is preferable to use this method. The West–Gaeke spectrophotometric method is the standard method for SO_2 monitoring of 0.0005-5 ppm SO_2 in the ambient air.

2.2 Air quality index determination

Based on the monitoring data of various pollutants at different sites on different days, an index called "Air Quality Index" was calculated using the following equation, thereby assessing the present air quality status of Delhi. As per the EPA guidelines, pollutants such as O_3, CO, SO_2, NO_2, PM_{10}, and $PM_{2.5}$ are used for calculating the AQI for an area.

$$lp = \frac{l_{Hi} - l_{Lo}}{BP_{Hi} - BP_{Lo}}(Cp - BP_{Lo}) + l_{Lo} \tag{1}$$

lp=the index for pollutant P
Cp=the rounded concentration of pollutant P
BH_{Hi}=the breakpoint greater than or equal to Cp
LH_{i}=the AQI value corresponding to BP_{Hi}
l_{Lo}=the AQI value corresponding to BP_{Lo}
BP_{Lo}=the breakpoint less than or equal to Cp

In the present study, the pollutants used for calculating the AQI were NO_2, SO_2, and RSP. AQI was calculated for individual pollutants simultaneously at a place, and the maximum value corresponding to one particular pollutant was reported as the AQI of that area. The same procedure was adopted at all sites on each day, with AQI finally reported along with the responsible pollutants.

3. Results and discussion

For all sites, we have calculated the mean, standard deviation, and maximum, and we also calculated the geometric means. The geometric mean is the basis of preparing the air quality standards. The mean, standard deviation, maximum value, and geometric mean values are listed as below:

	RSP	FSP	NRSP	TSP	$PM_{2.5}/PM_{10}$	RSP/TSP
OKHLA						
GEO MEAN	268.6	126.71	405.35	687.71	0.47	0.39
MEAN	271.11	129.7	425.2	696.3	0.49	0.40
STD DEV.	39.1	28.6	139.1	117.4	0.11	0.11
MAXIMUM	320.3	157.3	599.2	827.6	0.56	0.53
IIT						
GEO MEAN	202.1	139.02	289.0	492.1	0.69	0.41
MEAN	204.3	144.1	290.7	495.0	0.71	0.41
STD DEV.	35.4	38.6	32.9	59.3	0.20	0.04
MAXIMUM	274.7	170.6	326.7	601.3	0.91	0.46
VV						
GEO MEAN	256.9	152.58	251.3	514.6	0.59	0.50
MEAN	260.7	159.5	260.3	521.1	0.65	0.50
STD DEV.	49.7	45.0	77.9	97.3	0.26	0.09
MAXIMUM	322.6	198.1	394.5	717.2	0.94	0.64
SKP						
GEO MEAN	226.8	174.05	192.3	427.6	0.77	0.53
MEAN	233.0	175.0	199.4	432.4	0.78	0.54
STD DEV.	64.5	21.2	67.1	73.7	0.18	0.11
MAXIMUM	321.2	198.4	299.7	497.4	0.95	0.66
Delhi						
GEO MEAN	238.1	144.8	283.3	532.0	0.61	0.45
MEAN	243.1	150.0	302.5	545.6	0.65	0.46
STD DEV.	51.1	37.0	117.6	130.3	0.21	0.10
MAXIMUM	322.6	198.4	599.2	827.6	0.95	0.66
ITO Delhi						
GEO MEAN	245.65		371.35	628.99		0.39
MEAN	257.62		384.62	642.24		0.40
STD DEV.	86.28		101.04	130.81		0.10
MAXIMUM	586.00		659.00	984.00		0.63

Table 1. Statistical Analysis of Results.

3.1 General discussion

In general, for $PM_{2.5}$ the secondary formation is more relevant than the primary emissions, but Wilson and Suh (1997) stated that some natural particles also occur in size range <2.5μm. A typical study (Pekkanen et al., 1997) reports that a considerable fraction of the road dust consisted of particles<2.5μm in diameter. The primary particles are emitted by a source as particles and are dispersed in the atmosphere without any major chemical transformation (Shandilya and Kumar, 2010a, 2010b, 2011).

PM_{10} mainly consisted of three distinct components: Primary vehicular emissions, secondary pollutants, and windblown dust. Specifically, major contributors to excessive PM_{10} levels were combinations of area-wide emitters such as fugitive dust from roads, construction, and

agriculture; smoke from residential wood combustion or prescribed burning; directly emitted exhaust from motor vehicle engines; and secondary sulfates, nitrates, and organics formed from gaseous ammonia, sulfur dioxide, nitrogen oxides, and reactive organic gas emissions (Sharma et al., 2004, Shandilya et al., 2012). The emissions inventories for these "nontraditional" types of sources were found to be much less accurate than those obtained for "traditional" ducted emissions from the industrial sources. The coarse fraction was mainly composed of the re-suspended dust. Farming operations (e.g., agriculture tillage, harvesting, and travel on unpaved roads) account for large portions of the primary PM_{10} in many emissions inventories. PM_{10} non-attainment areas are suspected of having RWC as a major contributor. The prescribed burning of agricultural fields and forest slash is suspected of being a major contributor to PM_{10} in other areas. PM_{10} is considered more a local and urban scale problem, whereas $PM_{2.5}$ has a much longer residence time and is considered a regional or trans-boundary problem. The spatial and temporal distribution of fine particle levels may vary substantially.

Natural background levels, excluding all anthropogenic sources in the US and elsewhere, were 4-11-μgm^{-3} and 1-5-μgm^{-3} for PM_{10} and $PM_{2.5}$, respectively. PM_{10} mean concentrations were typically from 10-80-μgm^{-3} around the world. 10-20-μgm^{-3} is found at remote sites with no local sources (i.e., even in clean air). In urban areas, 60–220-$\mu g/m^3$ is typical, and in a heavily polluted area, levels may approach 2000-$\mu g/m^3$. More than 50-μgm^{-3} was found at heavily polluted sites with high traffic density and/or local sources in the surroundings, where heavily polluted urban areas may reach 100-μgm^{-3} on average. In the heavily polluted cities of southern Europe, Latin America, and Asia, average PM_{10} concentrations have reached 100-μgm^{-3}, and more. $PM_{2.5}$ mean concentrations were between 7-80-μgm^{-3}. PM_{10} trends were decreasing in the last decade, for emission and ambient levels, in the developed and developing world. The ratio of $PM_{2.5}$ to PM_{10} at the urban sites usually averaged 0.55-0.6.

Many studies indicated that TSP and PM_{10} concentration in the ambient air would be affected by various meteorological factors such as wind speed, wind direction, solar radiation, relative humidity, rainfall, and source conditions. Apart from the emission factors of the vehicles, SPM concentrations on the street level would be mainly affected by the mechanical turbulence (created by moving vehicles and wind) and by thermal turbulence produced by the hot vehicle exhaust gas (Shandilya and Kumar, 2009) .

24-hour average concentrations of the FSP, RSP, and TSP were calculated (See Methodology for a sample calculation) from samples collected at four stations at Southern Delhi and East Delhi and from samples taken from Satna. Our study is also not limited by the fact that, during winter, mixing depth in the early morning is lower than in the summer, thereby concentrating primary crustal aerosol emissions (e.g., from road dust). Since our study did not take place in the summer or winter, this condition is not applicable and the purpose of our study is not forfeited.

3.2 Comparison of SPM level in Delhi

The sources prevalent at VV are Diesel Buses, CNG Buses, Traffic, and Mechanical Disturbances, while at IIT the sources are Traffic, Vegetative Burning, construction work, and residential sources like cooking. At Okhla, the sources are Vegetative Burning, Diesel Generators, Mechanical Disturbances, Traffic, and Industrial Activities. Concentrations of

fine particles are close to main roads. Road transport and diesel exhaust are major sources of fine particles in urban areas (Shandilya and Kumar, 2009, Kumar et al., 2011). PM_{10} and $PM_{2.5}$ concentrations are elevated close to main roads compared to background sites. More monitoring details can be found in Shandilya et al (2006, 2011).

3.2.1 Variation of PM$_{2.5}$

With a minimum of one daily sample at a site, 24-hour $PM_{2.5}$ concentration at the urban-industrial Delhi site varies from 85.1-157.3μgm^{-3}. 24-hour $PM_{2.5}$ concentration at the urban-commercial Delhi site varies from 78.6-198.1μgm^{-3}, and 24-hour $PM_{2.5}$ concentration at the urban-residential Delhi sites varies from 84.6-198.4μgm^{-3}. The value at Delhi is well above 24-hour $PM_{2.5}$ USNAAQS (65μgm^{-3}). More details of this monitoring can be found in Shandilya et al., (2011). Variation of $PM_{2.5}$ value is highest on SKP, which suggests that the contribution from different sources varies to a great extent. The Standard Deviation values suggest that $PM_{2.5}$ source emission rate at VV varies greater than the emission rate at any place in South Delhi. The large variation is most likely due to the differences in air mass, emission characteristics, and meteorology.

3.2.2 Peak day composition

The maximum $PM_{2.5}$ concentration in Delhi, 198.4-$\mu g/m^3$, was at SKP on November 26, 2000, which may be due to a wash out caused by the rain. The lowest $PM_{2.5}$ value of 78.6-$\mu g/m^3$ was observed at VV bus depot (November, 24, 2000). When there was a strike and there was less traffic on the road, there was a significant decrease in $PM_{2.5}$ fraction. It shows an important relationship between $PM_{2.5}$ fractions with the traffic sources. Maximum $PM_{2.5}$ concentration in South Delhi, 198.1-$\mu g/m^3$, was at VV on November 22, 2000. Maximum $PM_{2.5}$ concentration at Okhla, 157.3-$\mu g/m3$, was on October 21, 2000. Maximum $PM_{2.5}$ concentration at IIT, 170.6-$\mu g/m^3$, was on November 6, 2000.

3.2.3 Variation of PM$_{10}$

With a minimum of one daily sample at a site, 24-hour PM_{10} concentration at the urban-industrial Delhi site varies from 205.0-320.3-μgm^{-3}. 24-hour PM_{10} concentration at the urban-commercial Delhi site varies from 210.0-322.6-μgm^{-3}, and 24-hour PM_{10} concentration at the urban-residential Delhi sites varies from 174.5-321.2-μgm^{-3}. The PM_{10} concentration value was always greater than the 24-hour NAAQS (150-$\mu g/m^3$ in industrial and commercial areas and 100-$\mu g/m^3$ in residential areas).

3.2.4 PM$_{2.5}$/PM$_{10}$ ratio

The most common ratio of $PM_{2.5}/PM_{10}$ is 0.60, but the prevalence of the local sources, with either a dominating fine or coarse fraction, may shift this ratio. $PM_{2.5}/PM_{10}$ concentration at the urban-industrial Delhi site varies from 0.27-0.56 with an average±standard deviation of 0.47±0.11. $PM_{2.5}/RSP$ concentration at the urban-commercial Delhi site varies from 0.25-0.94 with an average±standard deviation of 0.59±0.26. $PM_{2.5}/RSP$ concentration at the urban-residential South Delhi sites varies from 0.43-0.91 with an average±standard deviation of 0.69±0.20. $PM_{2.5}/RSP$ concentration at the urban-residential East Delhi sites varies from 0.62-0.95 with an average±standard deviation of 0.77±0.18. This shows that at Okhla, $PM_{2.5}$

contribution to PM_{10} is 47%, which increases to 59% at VV, further increases to 69% at IIT and achieves its maximum (77%) at SKP. $PM_{2.5}/PM_{10}$ ratio standard deviation is highest (0.26) at the VV site, suggesting the heterogeneous contribution of secondary particle formation sources. It has been observed that a part of PM_{10} is $PM_{2.5}$ but not as much as in the case of IIT in South Delhi. The standard deviation ($PM_{2.5}/RSP$) low value at Okhla (0.11) shows that the contribution of $PM_{2.5}/RSP$ is more consistent compared to other sites, and $PM_{2.5}$ sources contributing to RSP remains the same. The ratios of $PM_{2.5}/PM_{10}$ are generally between 20 and 30%, with abnormal values from industrial particulate (both high-50% in Cynon- and low-17% in Port Talbot). The highest contribution of $PM_{2.5}/PM_{10}$ strongly shows that industrial sources are predominant in the SKP area. Comparison with higher ratios generally found in large urban areas (Birmingham-QUARG report) indicates that the finer traffic particulate stick together or deposit on surfaces less rapidly than the coarse fraction of PM_{10}, so they tend to build up in larger urban areas or still conditions. It has been observed that a part of PM_{10} is $PM_{2.5}$, but not as much as in the case of SKP. It shows that the condensation products of the chemical reactions primarily contribute PM_{10} fraction that originated in this area. Data ($PM_{2.5}/PM_{10}$) shows that PM_{10} fraction that originated in industrial areas contains the condensation products of the chemical reactions and some primary particulate.

3.2.5 Peak day composition

The maximum PM_{10} concentration in Delhi, South Delhi and VV 322.6-$\mu g/m^3$ was on November 17, 2000 during our study; the lowest PM_{10} of 174.5-$\mu g/m^3$ was observed at Shakarpur. This can be due to some change of wind pattern since there is a reduction in $PM_{2.5}$ concentration (171.8-$\mu g/m^3$), which eradicates the possibility of contribution from secondary particles and is further confirmed by an increase in NRSP fraction. Maximum PM_{10} concentration at Okhla, 320.3-$\mu g/m^3$, was on October 29, 2000 next to Deepawali. This high concentration can be attributed to crackers; this is well confirmed by the reduction of $PM_{2.5}$ concentration (85.1-$\mu g/m^3$), which is primarily caused by vehicles and is further confirmed by decline in the contribution of $PM_{2.5}$ to PM_{10} suggested by the value of $PM_{2.5}/PM_{10}$ (0.27, second lowest in all). Maximum PM_{10} concentration at IIT, 274.7-$\mu g/m^3$, occurred on November 3, 2000, the day when a celebration was held, so there was a great rush of vehicles in IIT. The increase in PM_{10} was due to the resuspension of dust and vehicle exhaust. It was further confirmed by the increase in NRSP fraction on that day and an increase in $PM_{2.5}$. The $PM_{2.5}$ value on the second day remained on the same level (168.7-μgm^{-3}), showing that the residence time $PM_{2.5}$ was much higher, which was not the case with PM_{10} and NRSP. Maximum PM_{10} concentration at SKP, 321.2-$\mu g/m^3$, took place on November 26, 2000, which may have been due to the wash out caused by the rain.

3.2.6 Variation of TSP

With a minimum of one daily sample at a site, 24-hour TSP concentration at the urban-industrial Delhi site varies from 553.4-827.6-μgm^{-3}. 24-hr TSP concentration at the urban-commercial Delhi site varies from 453.2-717.2-μgm^{-3} and 24-hr TSP concentration at the urban-residential Delhi sites varies from 345.2-601.3-μgm^{-3}. The TSP concentration value was always greater than the 24-hr NAAQS of TSP i.e., 200-$\mu g/m^3$ in the residential area. The TSP concentration value was always greater than the NAAQS of TSP i.e., 500-$\mu g/m^3$ in the industrial and commercial area while it crossed the limit (NAAQS, 500-$\mu g/m^3$) only once on November 17, 2000 at the VV commercial area.

3.2.7 RSP/TSP ratio

RSP/TSP at the urban-industrial Delhi site varies from 0.27-0.56 with an average±standard deviation of 0.39±0.11. RSP/TSP concentration at the urban-commercial Delhi site varies from 0.42-0.64μgm^{-3} with an average±standard deviation of 0.50±0.09. RSP/TSP concentration at the urban-residential South Delhi sites varies from 0.37-0.46 with an average±standard deviation of 0.41±0.04. RSP/TSP concentration at the urban-residential East Delhi sites varies from 0.40-0.66 with an average±standard deviation of 0.53±0.11. This shows that at Okhla, RSP contribution to TSP is 39%, increasing to 50% at VV, decreasing to 41% at IIT, and again achieving its highest of 53% at SKP. The standard deviation (RSP/TSP) low value at IIT (0.04) shows that the contribution of RSP to TSP is consistent, while it varies equally at SKP and at Okhla. Therefore, the RSP emitting sources in the SKP area were contributing equally throughout the study, while these sources' emission pattern was changed at the other places.

3.2.8 Peak day composition

The maximum TSP concentration in Delhi, South Delhi and at Okhla 827.6-μg/m^3 occurred on October 23, 2000. The maximum TSP concentration at IIT, 601.3-μg/m^3, occurred on November 3, 2000. It is further confirmed by an increase in NRSP fraction on that day. The maximum TSP concentration at VV, 717.2-μg/m^3, occurred on November 17, 2000, which may be due to some change of wind pattern since there is a reduction in PM$_{2.5}$ concentration (171.8-μg/m^3) while there is an increase in NRSP fraction. The maximum TSP concentration at SKP, 489.4-μg/m^3, was on November 26, 2000, which may be due to the wash out caused by the rain. In short, TSP concentrations at IIT and at SKP sites are lower than the other values observed in Delhi, but PM$_{2.5}$ concentrations are higher than those in the Okhla industrial location.

Thus, industrial emissions have considerable impact on the ambient non-respirable particulate matter concentrations, while residential emissions have considerable impact on PM$_{2.5}$ concentration. Similarly, PM$_{10}$ concentrations at the Okhla site are higher than the values found at any other site. This suggests that the industrial emissions contribute more to PM$_{2.5-10}$ fraction.

Our observations at Okhla in South Delhi showed that PM$_{2.5}$, RSP, and TSP concentrations were 126.71±28.6, 268.6±39.1, and 687.71±117.4-μgm^{-3}, respectively; IIT showed that PM$_{2.5}$, RSP, and TSP concentrations were 139.02±38.6, 202.1±35.4, and 492.1±59.3-μgm^{-3}, respectively; and VV showed that PM$_{2.5}$, RSP, and TSP concentrations were 152.58±45.0, 251.3±49.7, and 514.6±97.3-μgm^{-3}, respectively. As stated before, SKP in East Delhi showed that PM$_{2.5}$, RSP and TSP concentrations were 174.05.0±21.1, 226.8±64.5, and 427.6±73.6-μgm^{-3}, respectively.

Our results could be compared with the results of Kanpur, a study carried out by Sharma et al. (2003). They found at the background site, IIT Kanpur that PM$_{10}$ levels were 62±11.22-μg/m^3 and at Agricultural University, the residential area, PM$_{10}$ levels were 71.46-335.83-μg/m^3. At the commercial area Naveen Market, PM$_{10}$ levels were 114.22-634.68-μg/m^3, and at the industrial area, Lajpat Nagar, PM$_{10}$ levels were 221.77-650.73-μg/m^3. On comparison, it can be concluded that PM$_{10}$ levels in all areas (industrial, commercial, residential) found the same range at both places (Delhi and Kanpur).

Our results could also be compared with the results of Jaipur, a study carried out by Das et al. (1998). They found at the industrial site that TSP levels were 384.61-µg/m^3 and, in another study by Gajraj et al. (submitted) in the Vishwakarma industrial area, mean PM$_{10}$ concentration during May 1999 was 242-µg/m^3; during June 1999, the mean concentration was PM$_{10}$ (208.5-µg/m^3). Our TSP levels at Okhla are certainly higher than TSP levels in Jaipur, as obtained by Das et al. (1998).

3.2.9 Explanation of spatial variation

The terminal settling velocity is an important factor for the spatial variation of SPM (Willeke & Baron, 1993). For very fine particles, such as a particle of 0.1-µm d_{ae} setting velocity, is 8.65x10^{-5}-cms^{-1} for a particle of 1-µm 3.48x10^{-3} and for a particle of 10-µm in size 3.06x10^{-1}-cms^{-1}. Primary particles (Aitken particles <0.1-µm) and large particles (e.g.>10-µm) are expected to have a bigger spatial variability than the particles in the accumulation mode (0.1–1-µm). Coagulation (for very fine particles <0.01-µm) and gravitational settling (for particles>1-µm) are the underlying mechanisms, which cause spatial heterogeneity. The distribution of PM$_{2.5}$ might be uniform in a situation where the secondary formation is important; in cities, however, with large emissions from heavy duties (diesel exhaust), PM$_{2.5}$ can also exhibit significant spatial variation. Secondary aerosols that contribute to the accumulation mode (0.1-2-µm) show quite homogeneous spatial distribution. In general, small-scale spatial variations of PM$_{2.5}$ were described to be smaller than the spatial variations of PM$_{10}$ (Tuch et al., 1997). The spatial variation was small for PM$_{2.5}$ but larger for PM$_{10}$. Our results are in agreement with Monn (2001). With respect to the small-scale spatial variation in the urban areas, the largest variations occurred in the ultrafine (0.1-µm) and coarse mode (PM$_{10-2.5}$) (Burton et al., 1996). Ultrafine particle number counts have large spatial variations, and they are not well correlated to mass data, so spatial variation is much more reflected in PM$_{10}$ than in PM$_{2.5}$.

The spatial variation was small for PM$_{2.5}$ but larger for PM$_{10}$. Spatial correlations for PM$_{2.5}$ are found to be lower than 0.9 and around 0.6 for PM$_{10}$. The contribution of coarse particle fraction (PM$_{10-2.5}$) on PM$_{10}$ can be important. Conclusively, we can say that SPM is complex with respect to its sources in Delhi. With the exception of the geological material, PM$_{2.5}$ source contribution estimates are not similar to those for PM$_{10}$. A deep insight in the source contribution could be obtained if we consider emission factors of SPM from different sources.

3.3 Oxides of sulfur and nitrogen

In order to understand the sources, behavior, and mechanism of particle formation in the atmosphere, it is important to measure the precursors like SOx and NOx; this primarily contributes to the formation of the secondary particulate, which again constitutes a big fraction of total particulate matter in the atmosphere. This type of precursor characterization for the continental, marine background, and urban influenced aerosols has been reported from various sites throughout the world (Garg et al., 2001). 24-hour average concentrations of SO$_2$ and NO$_2$ (See Methodology) from the samples collected at four stations at Southern Delhi and East Delhi are given in the table below; they were calculated by taking the average of the day and night value.

For all sites, we have calculated the mean, standard deviation, and minimum and maximum values. We have also calculated the geometric means to show a homogeneous distribution of the data. Geometric mean concentrations are better representations of the data population as the elements are log-normally distributed. The geometric mean, standard deviation, maximum and minimum values, and geometric mean values are listed as below:

Okhla	Day Value	Night Value	Sox 24-hr	Day Value	Night Value	NOx 24-hr	Sox/NOx 24-hr	Sox/NOx Day	Sox/NOx Night
Geo.Mean	5.86	25.47	15.76	32.48	107.85	70.68	0.22	0.18	0.24
Mean	6.13	25.55	15.84	32.58	110.57	71.58	0.23	0.19	0.24
Std Dev	1.89	2.32	1.76	2.73	24.20	11.68	0.06	0.06	0.08
Maximum	8.42	30.12	18.45	36.89	136.62	84.87	0.35	0.28	0.42
Minimum	3.50	23.58	13.54	28.20	64.55	50.72	0.18	0.11	0.19
IIT									
Geo.Mean	4.30	14.45	9.40	24.56	57.29	40.95	0.23	0.18	0.25
Mean	4.37	14.49	9.43	24.72	57.83	41.27	0.23	0.18	0.25
Std Dev	0.83	1.21	0.81	3.06	8.67	5.67	0.02	0.03	0.03
Maximum	5.78	16.32	10.48	29.85	70.37	50.11	0.26	0.22	0.29
Minimum	3.21	12.96	8.45	21.00	47.13	35.12	0.20	0.14	0.23
VV									
Geo.Mean	5.48	26.67	16.12	37.86	97.48	68.36	0.24	0.14	0.27
Mean	5.51	26.97	16.24	38.21	99.98	69.09	0.24	0.15	0.28
Std Dev	0.64	4.38	2.15	5.53	24.71	11.07	0.04	0.02	0.06
Maximum	6.32	33.18	19.12	44.60	139.19	85.49	0.30	0.16	0.38
Minimum	4.90	20.65	12.81	31.05	68.45	54.55	0.20	0.11	0.22
SKP									
Geo.Mean	5.32	29.66	17.49	42.88	133.91	88.45	0.20	0.12	0.22
Mean	5.40	30.00	17.70	42.99	134.91	88.95	0.20	0.12	0.22
Std Dev	1.07	5.26	3.13	3.47	19.36	11.10	0.02	0.01	0.02
Maximum	6.56	36.84	21.70	46.92	162.02	104.47	0.21	0.14	0.24
Minimum	4.46	24.52	14.52	39.45	116.09	78.43	0.17	0.11	0.19
Delhi									
Geo.Mean	5.22	22.83	14.11	32.98	92.48	63.19	0.22	0.16	0.25
Mean	5.38	23.81	14.59	33.81	98.28	66.04	0.23	0.16	0.25
Std Dev	1.36	6.65	3.69	7.51	33.25	19.08	0.04	0.04	0.06
Maximum	8.42	36.84	21.70	46.92	162.02	104.47	0.35	0.28	0.42
Minimum	3.21	12.96	8.45	21.00	47.13	35.12	0.17	0.11	0.19
ITO									
Geo.Mean			17.30			78.42	0.22		
Mean			17.81			80.57	0.22		
Std Dev			4.40			18.83	0.04		
Maximum			28.00			126.00	0.30		
Minimum			11.00			42.00	0.10		

Table 2. Statistical Analysis of the Result Obtained

3.3.1 Sources of sulfur oxides and nitrogen oxides

The natural sources of nitrogen oxides are the geothermal activity and bacterial action. Natural sources include the soil microbes, vegetation, biomass, slum, burning, and lightning. Natural sources include the biological emissions (e.g. vegetation and soil microbes) and abiotic processes (e.g., geogenic, lightning, and biomass burning). Globally, lightning strikes occur about 100-times/second (Borucki and Chameides, 1984). Lighting strikes are characterized by a highly ionized and high temperature channel, in which molecular nitrogen is dissociated and reacts with oxygen to form NO (Guenther et al, 2000).

The primary sources of NOx are the motor vehicles, electric utilities, and other industrial, commercial, and residential sources that burn fuels. Other sources include the chemical and nitric acid manufacture, the detonation of nitrate-containing explosives, and electric sources. Man made sources of nitrogen oxide include the automobile, railways (coal fired and diesel locos), airways (landing and takeoff operations), industrial processes like chemicals (nitric acid plants, processes where nitrogenous compounds are used), metal processing, fertilizer (nitrogenous fertilizers), and other miscellaneous activity like domestic combustion, forest fires and managed fires, industrial boilers, furnaces, and commercial combustion. Although most NO is produced by the natural bacterial action, it must be emphasized that the rate of emission from man-related sources has, by estimate, increased by one-fourth between 1966 and 1968. Certainly, man's greatest contribution to the family of nitrogen oxides arises from the combustion of fuels. The contribution to the total NOx can be categorized as Utilities 27%; Motor Vehicles 49%; Other Sources 5%; and Industrial/ Commercial/Residential 19%. Nearly 2/3 of NOx emissions originate at the Earth's surface from fossil fuel and biomass combustion. Vehicle exhausts are known to increase the inputs of NO_2 to the atmosphere (see, e.g., Hargreaves et al., 2000). The burning of natural gas (which produces negligible SOx, CO, and HC) does produce a very significant quantity of oxides of nitrogen.

Man caused nitrogen emissions as equivalent NO_2 are estimated as 53×10^6 TPA over the world, 95% of which are emitted in the Northern Hemisphere. Half of these are from the combustion of coal. Natural sources of NOx are estimated as between 15 and 17-times pollutant emissions. (Sawyer et al, 1998). The major source of NOx is the combustion when the fixation of atmospheric N occurs at the high flame temperature. The oxides are emitted mainly as NO, which is normally rapidly oxidized to NO_2 by atmospheric O_2 and O_3. Motor vehicle exhaust contributes a sizable fraction of the total emissions of NOx. As well as the stationary combustion sources, the manufacture of HNO_3 and nitrate fertilizer is the source of NO & NO_2. On a global basis, natural emissions of oxides of nitrogen (such as releases from soil and the ocean, lightning strikes, oxidation of ammonia, and stratospheric oxidation) all make substantial contributions to the total emissions.

Garg et al (2001) has reported the sectoral shares of NOx emissions for India in 1995. According to them, a major part of NOx emissions is contributed from 28% by the non-energy sources, 28% by electric power generation, 2% by road transport, 19% by Biomass burning, 4% by other transports, and 19% by industries. Delhi tops the table of top 10 NOx emitting districts in the terms of total emissions by emitting total NOx 66-Gg in 1990 and 82-Gg in 1995; in the terms of emissions per unit area, NOx/area (tons km^{-2}) was 44 in 1990 and 55 in 1995. The largest NOx emission chunk in Delhi is from the oil sources.

The major atmospheric man made sources of sulphur dioxide (SO_2) are the burning of fossil fuel in stationary combustion and industrial process, viz. petroleum, chemical, metallurgical and mineral industries, domestic emission and mineral industries, domestic emission, and fires, while the natural source are the volcanoes (wherever these are present, but in India there are none), geothermal activity, the bacterial decomposition of organic matter, forest fires, and managed fires. Motor vehicles are a relatively minor source of SO_2, since refined motor fuel normally has low sulfur content.

Ambient SO_2 results largely from the stationary sources (that burn the coal and oil, refineries, pulp, and paper mills) and from the nonferrous metal smelters. The sources of the pollution include the emissions from the on-road vehicles, non-road vehicles (like planes, ships, trains and industries), and small businesses and households where polluting products are used.

Nitrogen oxides (NOx) are produced when fossil fuels are burned in the motor vehicles, power plants, furnaces, and turbines. Other gasoline powered mobile sources (motorcycles, recreational vehicles, lawn, garden, and utility equipment) have high emissions on per quantity of the fuel consumed basis, but their contribution to the total emissions is small. Heavy-duty diesel vehicles are the dominant mobile source of the Oxides of nitrogen. Oxides of nitrogen emissions on a fuel-consumed basis are much greater from the diesel mobile sources than from the gas.

Another common source of SO_2 in the atmosphere is the metallurgical operations. Many ores, like Zn, Cu, and Pb are primarily sulfides. During the smelting of these ores, SO_2 is evolved in the stack concentrations of 5-10% (SO_2). Among miscellaneous operations releasing SO_2 into the atmosphere are the sulphuric acid plant, paper-manufacturing plants, open burning of the refuse, and municipal incinerators which contribute some amount of SO_2 to the atmosphere (Peavy et al, 1985).

In the entire sulphur entering atmosphere, 1/3rd is by the anthropogenic activities, and 2/3rd come from natural sources such as Hydrogen Sulfide (H2S) or SOx. The problem with the manmade sources is the distribution, rather than the amount. Delhi, in this instance, is being recognized as a city with a potentially high traffic load on its roads. Therefore, the combustion of fossil fuels like diesel and petrol can be considered as a major source of the SO_2 in Delhi. Besides this thermal power plant, which is located in the Southeast about 1-1.5 km far from the SKP station, it is also contributing SO_2 in the atmosphere because the coal is used as a main fuel for the heat and the power generation. However, SO_2 concentration in the air also depends upon the sulfur content of fuels that vary from less than 1% of good quality anthracite to over 4% for bituminous coal.

Most crude petroleum products contain less than 1% sulfur; a few contain up to 5%. In the coming years, Delhi is supposed to have less concentration of SO_2 in its atmosphere because the fuel with less S content has already been introduced into the market. Another common source of the sulfur dioxide in Delhi is metallurgical operation. Among the miscellaneous operations releasing releasing SO_2 into the atmosphere are sulfuric plants and paper manufacturing plants. The open burning of refuse and municipal incinerator also contribute some amount of SO_2.

Garg et al (2001) also reported the sectoral shares of SO_2 emissions for India in 1995. According to them, a major part of SO_2 emissions is contributed to 46% by non-energy

sources, 36% by electric power generation, 8% by road transport, 6% by biomass burning, 3% by other transports, and 1% by industries. The details of industrial SO_2 emissions shows that 40%, 27%, 14%, 10%, 7%, and 2% are shared by non-energy industries, steel industry, fertilizer, cement, other industries, and refineries, respectively.

In Delhi, SO_2/area (tkm^{-2}) in 1990 was 47.9, in 1995 it was 46.7, and the average annual concentrations (µg/m^3) were 16 in 1990 and 24 in 1995. The largest contribution to SO_2 levels is by the transport sector. Our study results should be considered by keeping the points of the study done by Kamyotra et al (2000) and Jain (2001). These studies have pointed out two important points: by increasing the sampling period, the concentration shown becomes less (Kamyotra et al., 2000) and the diurnal pattern over 24 hours changes with time, local conditions, and activities (Jain, 2001).

The nighttime high values can be attributed to the phenomenon of the inversion. According to Kamyotra et al (2000), the lower values found during the day can be ascribed for the evaporation of the liquid, but this evaporation is absent during the night time which, in turn, increases the night time values, so high values obtained can be attributed to the combined effect of these facts.

3.3.2 Variation in SO_2

With a minimum of one 12-hour daily sample at a site, 12-hour a day SO_2 concentration at the urban-industrial Delhi site varies from 3.50-8.42-µgm^{-3}, 12-hour a night SO_2 concentration varies from 23.58-30.12-µgm^{-3}, and the average 24-hour SO_2 concentration varies from 13.54-18.45-µgm^{-3}. With a minimum of one 12-hour daily sample at a site, 12-hour a day SO_2 concentration at the urban-commercial Delhi site varies from 4.90-6.32-µgm^{-3}, 12-hour a night SO_2 concentration varies from 20.65-33.18-µgm^{-3}, and average 24-hour SO_2 concentration varies from 12.81-19.12-µgm^{-3}. With a minimum of one 12-hour daily sample at two sites, 12-hour a day SO_2 concentration at the urban-residential Delhi site varies from 3.21-6.56-µgm^{-3}, 12-hour a night SO_2 concentration varies from 12.96-36.84-µgm^{-3}, and average 24- hour SO_2 concentration varies from 8.45-21.70-µgm^{-3}.

On Okhla, the highest day SO_2 concentration variation (1.89-µgm^{-3}) suggests that the source emission rate variation at Okhla is higher comparatively. On the VV site, day SO_2 concentration variation was lowest in all (0.64-µgm^{-3}), suggesting that the source diversity and source emission rate variation are lower comparatively. On the IIT site, day SO_2 concentration variation is 0.83-µgm^{-3}, and on the SKP site day SO_2 concentration variation is 1.07-µgm^{-3}. In all of Delhi, day SO_2 concentration varies from 3.21-8.42-µgm^{-3}.

On SKP, Night SO_2 concentration variation was the highest in all (5.26-µgm^{-3}), suggesting that the source emission rate variation at SKP is higher comparatively. On the IIT site, night-SO_2 concentration variation was the lowest in all (1.21-µgm^{-3}), suggesting that the source diversity and source emission rate variation are lower comparatively. On the VV site, night SO_2 concentration variation is 4.38-µgm^{-3}, and on the Okhla site night SO_2 concentration variation is 2.32-µgm^{-3}. In all of Delhi, night SO_2 concentration varies from 12.96-36.84-µgm^{-3}. On SKP, 24-hour SO_2 concentration variation was highest in all (3.13-µgm^{-3}), suggesting that the source emission rate variation at SKP is higher comparatively. On the IIT site, 24-hour SO_2 concentration variation was the lowest in all (0.81-µgm^{-3}), suggesting that the source diversity and source emission rate variation are lower comparatively. On the VV site,

24-hour SO_2 concentration variation is 2.15-μgm^{-3}, and on the Okhla site 24-hour SO_2 concentration variation is 1.76-μgm^{-3}. In all of Delhi, 24-hour SO_2 concentration varies from 8.45-21.70-μgm^{-3}.

3.3.3 Peak day composition

The maximum day SOx concentration in Delhi, 8.42-$\mu g/m^3$, was at Okhla on October 25, 2000. The lowest day SOx value 3.21-μgm^{-3} was observed at IIT on November 2, 2000. The maximum day SOx concentration in SKP, 6.56-$\mu g/m^3$, was on November 27, 2000. The maximum day SOx concentration at VV, 6.32-$\mu g/m^3$, was on November 22, 2000. The maximum night SOx concentration in Delhi, 36.84-$\mu g/m^3$, was at SKP on November 27, 2000. The lowest night SOx value 12.96-μgm^{-3} was observed at IIT on November 5, 2000. The maximum night SOx concentration in Okhla, 30.12-$\mu g/m^3$, was on October 23, 2000. The maximum night SOx concentration at VV, 33.18-$\mu g/m^3$, was on November 23, 2000.

The maximum 24-hour SOx concentration in Delhi, 21.70-$\mu g/m^3$, was at SKP on November 27, 2000. The lowest 24-hour SOx value, 8.45$\mu g/m^3$, was observed at IIT on November 2, 2000. The maximum 24-hr SOx concentration in Okhla, 18.45-$\mu g/m^3$, was on October 23, 2000. The maximum 24-hour SOx concentration at VV, 19.12-$\mu g/m^3$, was on November 23, 2000. Summarizing our observations at Okhla in South Delhi showed that day SOx, night SOx, and average 24-hour concentrations were 5.86±1.89, 25.47±2.32, and 15.76±1.76-μgm^{-3}, respectively. IIT showed that day SOx, night SOx, and average 24-hour concentrations were 4.30±0.83, 14.45±1.21, and 9.40±0.81-μgm^{-3}, respectively. VV showed that day SOx, night SOx, and average 24-hour concentrations were 5.48±0.64, 26.67±4.38, and 16.12±2.15-μgm^{-3}, respectively. SKP in East Delhi showed that day SOx, night SOx, and average 24-hour concentrations were 5.32±1.07, 29.66±5.26, and 17.49±3.13-μgm^{-3}, respectively, and overall in Delhi were 5.22±1.36, 22.83±6.65, and 14.11±3.69-μgm^{-3}.

3.3.4 Variation in NO_2

With a minimum of one 12-hour daily sample at a site, 12-hour per day NO_2 concentration at the urban-industrial Delhi site varies from 28.20-36.89-μgm^{-3}, 12-hour per night NO_2 concentration varies from 64.55-136.62-μgm^{-3} and average 24-hour NO_2 concentration varies from 50.72-84.87-μgm^{-3}. With a minimum of one 12-hour daily sample at a site, 12-hour per day NO_2 concentration at the urban-commercial Delhi site varies from 31.05-44.60-μgm^{-3}, 12-hour per night NO_2 concentration varies from 68.45-139.19-μgm^{-3}, and average 24-hour NO_2 concentration varies from 54.55-85.49-μgm^{-3}. With a minimum of one 12-hour daily sample at two sites, 12-hour per day NO_2 concentration at the urban-residential Delhi site varies from 21.00-46.92-μgm^{-3}, 12-hour per night NO_2 concentration varies from 47.13-162.02-μgm^{-3} and average 24-hour NO_2 concentration varies from 35.12-104.47-μgm^{-3}.

On VV, day NO_2 concentration variation was highest in all (5.53-μgm^{-3}), which suggests that the source emission rate variation at VV is higher comparatively. On the Okhla site, day NO_2 concentration variation was the lowest in all (2.73-μgm^{-3}), which suggests that the source diversity and source emission rate variation is lower comparatively. On the IIT site, day-NO_2 concentration variation is 3.06-μgm^{-3}, and on the SKP site, the day NO_2 concentration variation is 3.47-μgm^{-3}. In all of Delhi, the day NO_2 concentration varies from 21.00-46.92-μgm^{-3}.

On the VV site, night-NO_2 concentration variation was the highest in all (24.71-$\mu g/m^3$), which suggests that the source emission rate variation at SKP is higher comparatively. On the IIT site, night NO_2 concentration variation was the lowest in all (8.67-μgm^{-3}), which suggests that the source diversity and source emission rate variation is lower comparatively. On the Okhla site night NO_2 concentration variation is 24.20-μgm^{-3}, and on the SKP site night NO_2 concentration variation is 19.36-μgm^{-3}. In all of Delhi, night NO_2 concentration varies from 47.13-162.02-μgm^{-3}.

On Okhla, 24-hour NO_2 concentration variation was highest in all (11.68-μgm^{-3}), which suggests that the source emission rate variation at Okhla is higher comparatively. On the IIT site, 24-hour NO_2 concentration variation was the lowest in all (5.67-μgm^{-3}), which suggests that the source diversity and source emission rate variation are lower comparatively. On the VV site, 24-hour NO_2 concentration variation is 11.07-μgm^{-3}, and on the SKP site, 24-hour NO_2 concentration variation is 11.10-μgm^{-3}. In all of Delhi, 24-hour NO_2 concentration varies from 35.12-104.47-μgm^{-3}.

3.3.5 Peak day composition

The maximum day NOx concentration in Delhi, 46.92-$\mu g/m^3$, was at SKP on November 27, 2000. The lowest day NOx value, 21.00-$\mu g/m3$, was observed at IIT on November 4, 2000. The maximum day NOx concentration in Okhla, 36.89-$\mu g/m^3$, was on October 25, 2000. The maximum day NOx concentration at VV, 44.60-$\mu g/m^3$, was on November 15, 2000. The maximum day NOx concentration at IIT, 29.85-$\mu g/m^3$, was on November 6, 2000.

The maximum night NOx concentration in Delhi, 162.02-$\mu g/m^3$, was at SKP on November 27, 2000. The lowest night NOx value, 47.13-μgm^{-3}, was observed at IIT on November 2, 2000. The maximum night NOx concentration in Okhla, 136.62-$\mu g/m^3$, was on October 23, 2000. The maximum night NOx concentration at VV, 139.19-$\mu g/m^3$, was on November 23, 2000. The maximum night NOx concentration at IIT, 70.37-$\mu g/m^3$, was on November 6, 2000.

The maximum 24-hour NOx concentration in Delhi, 104.47-$\mu g/m^3$, was at SKP on November 27, 2000. The lowest 24-hour NOx value, 35.12-μgm^{-3}, was observed at IIT on November 2, 2000. The maximum 24-hour NOx concentration in Okhla, 84.87-$\mu g/m^3$, was on October 23, 2000. The maximum 24-hour NOx concentration at VV, 85.49-$\mu g/m^3$, was on November 23, 2000. The maximum NOx concentration at IIT, 50.11-$\mu g/m^3$, was on November 6, 2000.

Our observations at Okhla in South Delhi showed that day NOx, night NOx, and average 24- hour concentrations were 32.48±2.73, 107.85±24.20, and 70.68±11.68-μgm^{-3}, respectively. Observations at IIT showed that day NOx, night NOx, and average 24-hour concentrations were 24.56±3.06, 57.29±8.67, and 40.95±5.67-μgm^{-3}, respectively. VV showed that day NOx, night NOx, and average 24-hour concentrations were 37.86±5.53, 97.48±24.71, and 68.36±11.07-μgm^{-3}, respectively. SKP in East Delhi showed that day NOx, night NOx, and average 24-hour concentrations were 42.88±3.47, 133.91±19.36, and 88.45±11.10-μgm^{-3}, respectively, and the overall concentrations in Delhi were 32.98±7.51, 92.48±33.25, and 63.19±19.08-μgm^{-3}.

As evident from the table 2, the concentration of NOx was the highest at SKP, followed by Okhla, VV, and IIT. The trend is attributed to the prevailing activities at the sites. 24-hour average NOx concentration is mainly contributed by the nighttime values. These nighttime higher NOx concentrations at the different sites also reflect the local source conditions.

3.3.6 SOx to NOx day ratio

The day ratio at an urban-industrial Delhi site varies from 0.11-0.28 and at the urban-commercial Delhi site varies from 0.11-0.16; at two urban-residential Delhi sites, it varies from 0.11-0.22. At SKP, variation of the ratio (0.01) is lowest, while it is highest at the Okhla (0.06). The variation of ratio at VV was 0.02, and at IIT it was 0.03, and in Delhi the ratio ranged from 0.11-0.28. The variation in Delhi (4%) shows that the sources liberating NOx are also releasing SOx. Summarizing our observations at Okhla in South Delhi showed that the day ratio was 0.18±0.06. IIT in South Delhi showed that the ratio was 0.18±0.03. VV in South Delhi showed that the ratio was 0.14±0.02. SKP in East Delhi showed that the ratio was 0.12±0.01, and overall the Delhi ratio was 0.16±0.04.

3.3.7 SOx to NOx night ratio

The night ratio at the urban-industrial Delhi site varies from 0.19-0.42, and at the urban-commercial Delhi site varies from 0.22-0.38, and at two urban-residential Delhi sites varies from 0.19-0.29. At SKP, the variation of the ratio (0.02) is the lowest, while it remains the highest at Okhla (0.08). The variation of ratio at VV was 0.06, and at IIT it was 0.03, and in Delhi the ratio ranges from 0.19-0.42. The variation in Delhi (6%) shows that sources liberating NOx are also releasing SOx. Summarizing our observations at Okhla in South Delhi showed that the night ratio was 0.24±0.08. IIT in South Delhi showed that the ratio was 0.25±0.03. VV in South Delhi showed that the ratio was 0.27±0.06. SKP in East Delhi showed that the ratio was 0.22±0.02, and overall in Delhi the ratio was 0.25±0.06.

3.3.8 Sox to NOx 24-hr ratio

The 24-hour ratio at the urban-industrial Delhi site varies from 0.18-0.35, and at the urban-commercial Delhi site varies from 0.20-0.30, and at two urban-residential Delhi sites varies from 0.17-0.26. The SKP and IIT variation of the ratio (0.02) is the lowest, while the highest is at Okhla (0.06). The variation of ratio at VV was 0.04, and overall in Delhi the ratio ranges from 0.17-0.35. The variation in Delhi (4%) shows that the sources liberating NOx are also releasing SOx.

Summarizing our observations at Okhla in South Delhi showed that the 24-hour ratio was 0.22±0.06. IIT in South Delhi showed that the ratio was 0.23±0.02. VV in South Delhi showed that the ratio was 0.24±0.04. SKP in East Delhi showed that the ratio was 0.20±0.02, and overall in Delhi the ratio was 0.22±0.04.

Many studies indicated that NOx and SOx concentration in ambient air would be affected by the various meteorological factors such as the wind speed, wind direction, solar radiation, relative humidity, rainfall, and source conditions. Our results could also be compared with the values obtained by the CPCB personnel at ITO during the study. These were taken from their site (http://www.envfor.nic.in/). Our values are in good agreement with the CPCB values. During comparison, it was kept in mind that these values are of those samples, which were taken for eight-hour duration as per CPCB rules. But these values are normalized for 24-hours to represent the daily mean value (as per CPCB personnel).

The SOx levels were always lesser than the 24-hour NAAQS of SOx, i.e., 80-µg/m^3 in the residential area and 120-µg/m^3 in the industrial area. The NOx levels were lesser than 24-

hour NAAQS of NOx i.e. 80-µg/m³ in the residential area and 120-µg/m³ in the industrial area, except on November 23, 2000 at VV and on November 26, 27, and 28, 2000 at Shakarpur (80-µg/m³).

The ITO site can be considered as a commercial site in East Delhi, so we will compare our results of VV with it. We will also compare our results of SKP with it. The levels obtained at ITO in East Delhi showed that NOx, SOx concentrations and SOx/NOx ratio were 78.42±18.83, 17.30±4.40-µgm⁻³, and 0.22±0.04, while at VV levels were 68.36±11.07, 16.12±2.15-µgm⁻³, and 0.24±0.04. Our data also did not show any weekend or holiday effect. The variation is most likely due to differences in the air mass, emission characteristics, and the meteorology.

3.3.9 Comparison with other studies

Das et al (1997) reported in NO_2, SO_2, and NO_2/SO_2 levels in Jaipur for the sampling time of two-hours in the evening peak traffic hours at 49-stations. At residential areas, NO_2 and SO_2 values varied between 4.61-332.89-µg/m³ and 115.84-296.74-µg/m³; at the commercial areas, between 1.07-974.44-µg/m³ and 136.96-912.64-µg/m³; and at the industrial areas, between 26.16-78.81-µg/m³ and 141.44-262.02-µg/m³. Gajraj et al (2000) reported for May-June 1999 SO_2 and NOx levels during the morning and the evening for the rush hours in Jaipur as 56.7-184.4-µg/m³ and 70.8-139-µg/m³. Sharma et al. (2003) reported SO_2 levels in Kanpur for 24-hours at five stations. They found at IIT Kanpur SO_2, the levels were 5.44±1.58-µg/m³, at GT road were 28.6±9.9-µg/m³, at Agricultural University were 11.0±2.7-µg/m³, at Naveen Market were 14.6±3.8-µg/m³, and at Lajpat Nagar were 17.73±4.38-µg/m³.

CPCB (http://www.envfor.nic.in/divisions/cpoll/delpoll.html) have reported an increase in the annual SO_2 level over the years (1989-1997), such as 8.7, 10.2, 13.3, 18.4, 18.5, 19.5, 19.0, 19.0, and 16.2-µg/m³, and in annual NO_2 level such as 18.5, 22.5, 27.2, 30.4, 33.2, 33.0, 34.1, 33.7, and 33.0-µg/m³. Though the annual mean value of SO_2 (15-26-µg/m³) and NOx (28-46-µg/m³) remain within the prescribed limits of 60-80-µg/m³, there is a rising trend. Compared to 1989, SO_2 atmospheric concentrations in 1996 have registered a 109% rise, and NOx an 82% rise. Glikson et al (1995) reported in Brisbane, Australia for the period of the study that the maximum 1-hour average for NO_2 was 6.7-ppm. Cheng and Lam (2000) reported in Hong Kong the seasonal daily mean SO_2 concentrations for the entire period (1983-1992) were 28.2, 38.3, 20.0, and 23.5-µgm⁻³ for spring, summer, autumn, and winter, respectively; their standard deviations were 30.8, 42.7, 16.9, and 23.6-µgm⁻³. The corresponding figures of the overall mean NOx concentrations were 129.7, 110.4, 121.0, and 148.6-µgm⁻³ with standard deviations of 54.2, 49.7, 40.1, and 74.5-µgm⁻³. Mondal et al (2000) reported the ground level concentration of NOx at 19-important traffic intersection points in Calcutta ranging from 55-222-µgm⁻³. Lebret et al (2000) reported small area variations in ambient NO_2 concentrations in four European areas, i.e., in Amsterdam (Netherlands), Huddersfield (UK), Poznan (Poland), and Prague (Czech Republic) found NO_2 concentrations as 24-72, 10-79, 12-48, and 9.6-18.4-µgm⁻³. In NOx emission Delhi holds second position, and in SO_2 Delhi is not in the top 10-districts of India (Garg et al., 2001). India is the second largest contributor of SO_2 emissions in Asia, after China. For India, the estimates of national SO_2 emissions (Gg/yr.) in the period 1985-97 shows a rising trend as the emissions were 3402.0, 3609.1, 3829.9, 4085.2, 4256.1, 4437.2, 4684.3, 4863.1, 5039.6, 5289.3, 5609.5, 6106.3, and 6276.6-Gg/yr in the years 1985, 1986, 1987, 1988, 1989, 1990, 1991, 1992, 1993, 1994, 1995, 1996, and 1997, respectively (Streets et al., 2000). Our study is in good agreement with all of these studies.

3.4 Air quality index

In order to assess the degree of air pollution in Delhi, an index called air quality index (AQI) has been used as recommended by USEPA (EPA-454/R-99-010). More details are given in the literature review section, and the calculation method adopted is given in the methodology section. Calculated AQI has been presented in the table below.

DATE	RSP	SOx	NOx	AQI RSP	AQI SOx	AQI NOx
Okhla						
20/10	259.06	14.41	77.48	153.03	20.59	70.10
21/10	297.51	15.66	71.03	172.04	23.03	67.20
23/10	273.07	18.45	84.87	159.96	27.13	73.42
24/10	271.74	17.74	50.72	159.27	26.10	58.07
29/10	320.31	16.07	62.90	183.32	23.63	63.54
9-Nov	204.99	13.54	72.67	125.75	19.91	67.93
IIT						
2-Nov	179.09	8.45	35.12	112.93	12.43	51.05
3-Nov	274.65	9.09	42.62	160.75	13.37	54.53
4-Nov	189.11	9.57	36.17	117.88	14.07	51.53
5-Nov	196.80	8.76	38.94	121.69	12.88	52.77
6-Nov	200.86	10.22	50.11	123.72	15.03	57.79
7-Nov	185.06	10.48	44.69	115.90	15.41	55.36
VV						
15/11	245.62	12.81	63.90	145.84	18.84	63.99
16/11	258.31	17.68	62.80	152.63	26.00	63.50
17/11	322.63	15.26	77.01	184.46	22.44	69.89
22/11	210.57	16.34	70.80	128.52	24.03	67.09
23/11	209.97	19.12	85.49	128.23	28.12	73.70
24/11	317.32	16.24	54.55	181.84	23.88	59.79
SKP						
25/11	174.54	16.08	78.43	110.65	23.65	70.52
26/11	321.24	18.48	88.07	183.77	27.18	74.86
27/11	238.72	21.70	104.47	142.43	31.91	82.23
28/11	197.64	14.52	84.82	122.09	21.35	73.40

Table 3. Daily AQI along with Respective Parameter Concentrations

The category of air quality based on AQI for RSPM at Okhla was always unhealthy except on one day i.e. November 9, when it was unhealthy for sensitive people. The category of air quality based on AQI for RSPM at IIT was always unhealthy for sensitive people except on one day i.e. November 3, when it was unhealthy for the general public. The category of air quality

based on AQI for RSPM at VV was unhealthy on November 16, 17, and 24, 2000, while it was unhealthy for sensitive people on November 15, 22, and 23, 2000. The category of air quality based on AQI for RSPM at SKP was always unhealthy for sensitive people except on one day i.e. November 26, 2000, when it was unhealthy for the general public as well. The category of air quality based on AQI for SOx and NOx at all sites was always good for the public.

It can be concluded that PM is the critical parameter for the deterioration of ambient air quality in Delhi.

3.4.1 Comparison with other studies

In the industrial area of Jaipur, Gajraj et al (2000) found 24 observations of RSPM, where the air quality was unhealthy for 14 days and unhealthy for sensitive groups for the remaining days. For 32 observations of RSPM in Jaipur, Jain (2001) found that the air quality was good on six days, moderate on 20 days, and unhealthy for sensitive people on six days. She also found for 24 observations of SO2 that the air quality was good on all days.

4. Conclusions

The $PM_{2.5}$ value at Delhi is well above 24-hour $PM_{2.5}$ USNAAQS (65-μgm^{-3}). The variation of $PM_{2.5}$ value is highest on SKP, which suggests that the contribution from different sources is varying to a great extent. The PM_{10} concentration value was always greater than the 24-hour NAAQS (150-$\mu g/m^3$ in industrial and commercial areas and 100-$\mu g/m^3$ in residential areas). The study shows that, at Okhla, $PM_{2.5}$ contribution to PM_{10} is 47%; it increases to 59% at VV, further increases to 69% at IIT, and achieves its maximum (77%) at SKP. The highest contribution of $PM_{2.5}/PM_{10}$ strongly shows that industrial sources are predominant in the SKP area. This shows that at Okhla RSP contribution to TSP is 39%; it increases to 50% at VV and decreases to 41% at IIT and again achieves its highest of 53% at SKP. The above discussion put forward that the secondary formation and diesel exhaust, which are prime important mechanisms of the formation of $PM_{2.5}$, are predominant at IIT but the primary particulate sources also seem like prime important reasons for the formation of RSP. In all of Delhi, 24-hour SO_2 concentration varies from 8.45-21.70-μgm^{-3} and 24-hour NO_2 concentration varies from 35.12-104.47-μgm^{-3}. The variation in Delhi shows that sources liberating NOx are also releasing SOx. The SOx levels were always lesser than the 24-hour NAAQS of SOx i.e. 80-$\mu g/m^3$ in the residential area and 120-$\mu g/m^3$ in the industrial area. The NOx levels were lesser than the 24-hour NAAQS of NOx i.e. 80-$\mu g/m^3$ in the residential area and 120-$\mu g/m^3$ in the industrial area. The SO_2 and NO_x data do not show any weekend or holiday effect. The variations observed are most likely due to differences in the air mass, emission characteristics, and the meteorology. The category of air quality based on AQI for SOx and NOx at all sites was always good for the public. The category of air quality based on AQI for PM at all sites was always unhealthy for the public. It can be concluded that PM is the critical parameter for the deterioration of ambient air quality in Delhi.

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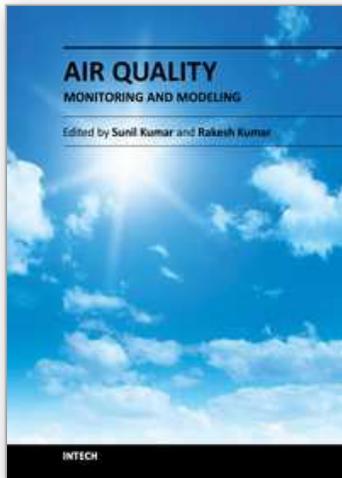
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