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A study of horizontal distribution pattern of particulate and gaseous pollutants based on ambient monitoring near a busy highway

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ABSTRACT

Understanding vehicular emissions, near-roadway pollutant dispersion, and their potential impact on near-roadway populations is an area of growing environmental interest in the developing countries. This paper presents horizontal distribution of coarse and fine particulate matter and gaseous pollutants in communities situated near the busy highway (NH-2) in the city of Agra. Over the total measured distance (0–500 m), the maximum decreases of NO_2, SO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$, $PM_{1.0}$ were found within 250 m distance from the highway of sites Khandari and Kaushalpur respectively. Annual average concentration for PM_{10} and $PM_{2.5}$ was found to be much higher than the standards prescribed by NAAQS and WHO, whereas levels of NO_2, O_3, NH_3 and SO_2 were somewhat within permissible limits. Significant seasonal variations were obtained with maximum concentration found in winter followed by summer and monsoon seasons. Higher values of winter/monsoon average ratios than the winter/summer average ratios of pollutants further signify the effect of meteorological conditions. The diurnal trend of pollutants clearly shows the effect of traffic density on the pollution level, as the peaks were obtained during the rush hours. Three factors have been identified using factor analysis (PCA) at 0 m, 250 m and 500 m distance from the highway. It showed that fine particles contributed a major part of PM_{10} with other gaseous pollutants at the roadsides, which is most likely attributed to the combinations of local sources including exhausted particulate matter from vehicles and re suspended fine dust and other variable sources.

1. Introduction

Road transport is globally recognized as a significant and constantly increasing source of air pollution. In some of the developed countries, like UK and USA, the problem of air pollution in urban areas has been successfully dealt with, while in most developing countries like China and India, the situation is rather serious due to their poor economy and insufficient financial investment for the control of urban air pollution (Zhang et al., 2000). Over some years there has been a rapid increase in urbanization and industrialization in many cities of India. With this has come a dramatic increase in the number and density of motor vehicles (Taneja et al., 2008). The increased traffic has resulted in increased pollutant emissions and the deterioration of environmental quality and human health in several major cities in India (Nagendra et al., 2007; Satsangi et al., 2016). Since motor vehicle emissions are a major source of air pollution, it has been suggested that urban residents living near roadways experience higher exposure to motor vehicle

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emissions compared to those living farther away, thereby experiencing higher health risk (Fujita et al., 1992; Derwent et al., 1995; Kodama et al., 2002; Zannoni et al., 2016).

Vehicles in major cities of India are estimated to account for 70% of the respective total pollution loads in these cities (Sharma et al., 2004; Apeagyei et al., 2011). Particulate matter pollution in these cities near major roads was often found to be more severe than that in urban areas since these are affected directly by various important primary sources. Resuspended road dust generally considered to be the most important source of PM, especially for the fraction of coarse matter (between 2.5 and 10 μm). Motor vehicle emissions, however, usually constitute a significant source of fine and ultrafine particles, such as $PM_{2.5}$ and $PM_{1.0}$ (Wu et al., 2002; Wagstrom and Pandis, 2011). Dockery and Pope (1994) reported that for each 10 $\mu g/m^3$ increase in the concentration of PM_{10} there is an estimate of increase in human mortality of 0.6–1.6%. The health impact of finest particulate ($PM_{2.5}$, $PM_{1.0}$) is greater because it can be inhaled deep into the unciliated and alveolar sections of the lungs (Massey et al., 2013). The high levels of NO_2 emitted from vehicle exhaust irritate the alveoli of lungs and also lead to increased airway resistance and decreased pulmonary diffusing capacity (Mudgal et al., 2000; Yang et al., 2005; Saini et al., 2008). It also acts as a precursor to ozone formation. Air pollution of surface ozone, arising from photochemical formation and accumulation, has plagued many regions worldwide (Greenfelt and Schjoldanger, 1984; Volz and Kley, 1988; Saini et al., 2008). SO_2 is usually emitted during the combustion of low-quality fossil fuels and one of the chief sources of fossil fuel pollution is the road traffic (Zou et al., 2007). High levels of SO_2 exposure have been related to increase in hospital admission for chronic bronchitis (Ciccone et al., 1995), low birth weights (Rogers et al., 2000) and reduction in birth rates (Dejmek et al., 2000).

Although a number of studies have sought to describe the distance decay relationship of pollutants near busy roadways (Roorda-Knape et al., 1998; Hitchins et al., 2000; Titta et al., 2002; Zhu et al., 2002a; Reponen et al., 2003; Zhu et al., 2006, Tsai et al., 2010, Squizzato et al., 2012; He and Dhaniyala, 2012; Wu et al., 2014), a few (Beckerman et al., 2008; Hong and Cheng, 2008; Kudo et al., 2011) have sought to assess the specific associations between pollutants. The relationship between human mortality and traffic related air pollutants were investigated (Tsai et al., 2010; Massey et al., 2016). However, there have been very few studies of local variation in ozone resulting from variation in vehicular traffic patterns within communities and of source identification of the pollutants along with the increasing distance from the highway. Therefore it is necessary to quantify the emission levels of the gaseous pollutants (NO_2, SO_2, O_3, NH_3) and particles with different size fraction (PM_{10}, $PM_{2.5}$, $PM_{1.0}$) near heavy traffic road and also to determine their behavior after emission as they are transported away from the road. Thus the present study is designed to measure the horizontal concentrations of particulate pollutants PM_{10}, $PM_{2.5}$, $PM_{1.0}$ and gases like NO_2, SO_2, O_3, NH_3 from the highway with the effect of meteorological parameters on them. The diurnal patterns of these pollutants were studied along with variation in vehicular traffic patterns within communities situated near the highway. Their probable sources were also identified responsible for their emission along with the increasing distance from the highway using the factor analysis. This study will help the city planners and the authority boards in most cities of the developing world, to know about the status of pollutants concentration adjacent to highways. Thus a proper understanding can be developed while planning and designing the communities adjacent to the busy highways.

2. Material and methods

2.1. Description of the sampling site

Agra, the city of the Taj, is located in north-central region (27°10′N 78° 83 02′E) of India. It is bounded by the Thar desert of Rajasthan on its South East, West and North West peripheries and is, therefore, a semi-arid area with a marked monsoon season (Fig. 1). Meteorologically, the year is divisible into three distinct seasons; summer, monsoon and winter. The summer season starts from March and extends up to June, monsoon season extends from July to September while winter season covers months from October to February. Summer is associated with strong hot dry westerly winds and high temperature ranges between 19.6 °C and 46.2 °C. Relative humidity in the summer ranges between 16.6% and 76.4%. The rainy season is hot and humid, temperature range from 22.5 °C to 37.9 °C and the relative humidity ranges between 57.2% and 89.0%. The pre monsoon and monsoon seasons are dominated by strong northeast and southeast winds, and in winter temperature ranges from 5.9 °C to 38.8 °C. Dust storms and thunderstorms are most commonly observed during the period from March to June. The wind speed varies from 0.1 to 19.8 km/h with highest values observed during summer and monsoon and lowest in winter. Table 1, shows the statistical data of meteorological parameters measured during the sampling period. Four busy highways, NH-2, NH-3, NH-11 and NH-19 cross the city. NH-2 is one of the busiest highways, which connects Delhi to Kolkata via Agra. On an average, on regular days, six to seven thousand vehicles move on this highway (Sharma et al., 2009). Agra city has around 1,585,704 total inhabitants and the district has a population density of about 1084 persons/km^2 (Census of India, 2011). The two sampling locations selected for the present study were Khandari (site A) and Kaushalpur (site B) which were lying perpendicular to the national highway (NH-2) as shown in Fig. 1. Khandari is a very busy crossing on NH-2 with residential colonies lying sideways while Kaushalpur is a densely populated residential area adjacent to NH-2 with high level of vehicular pollution, caused by the highway and localized traffic congestions. At each of the two locations, three sampling sites were set up at three different distances i.e. A1 (0 m), A2 (250 m), A3 (500 m) and B1 (0 m), B2 (250 m), B3 (500 m) from NH-2.

2.2. Sampling and chemical analysis

The present study was conducted from July 2009 to June 2010. The sampling period was divided up into winter

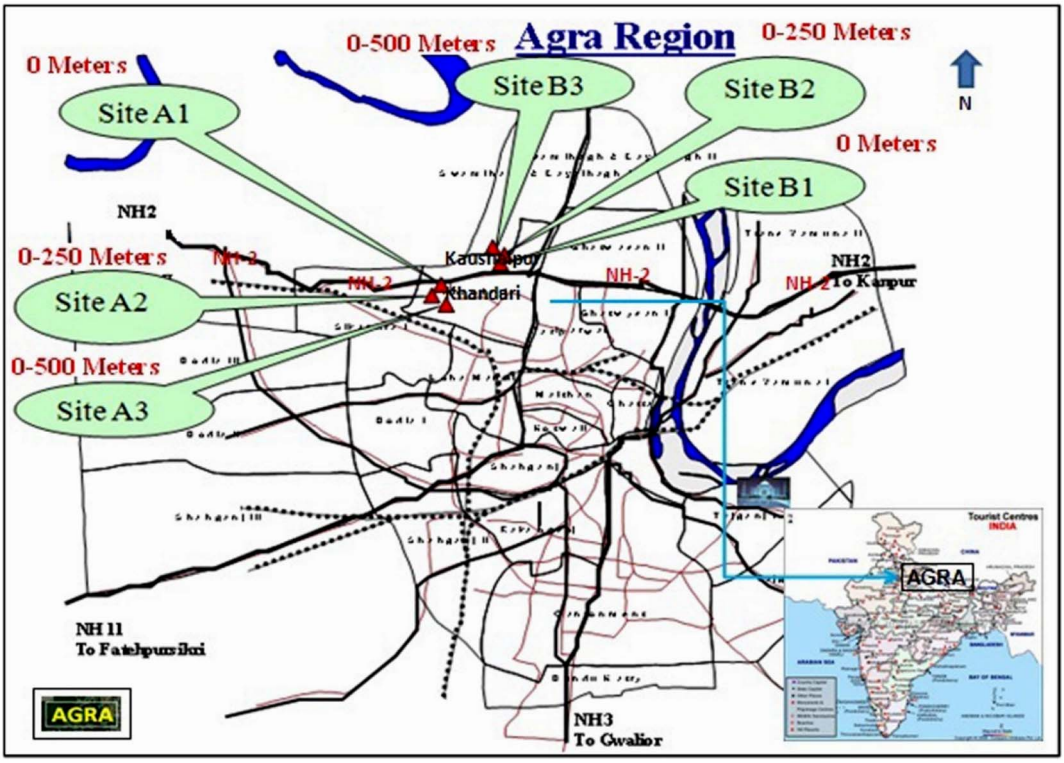

Fig. 1. Map of Agra showing various sampling sites near National Highway (NH-2).

Table 1
Description of meteorological conditions during different seasons of the sampling period.

Parameters	Summer	Monsoon	Winter
Temperature (°C)	53.49 ± 38.78	28.58 ± 3.09	20.32 ± 6.90
Relative humidity (%)	38.80 ± 13.06	77.54 ± 7.27	48.65 ± 17.88
Wind speed (km/S)	3.30 ± 2.58	2.62 ± 2.04	1.85 ± 2.08
Wind direction (°)	200.09 ± 19.80	210.74 ± 9.40	254.02 ± 18.75

(October–December 2009 and January–February 2010), summer (March–June 2010) and monsoon (July–September 2009). Monitoring for each pollutant was carried out for 288 days during the sampling duration (i.e. four times at each site in a month's duration). Sampling was conducted at the rate of 4 h for SO_2, NO_2, NH_3, 2 h for O_3 and 24 h for PM_{10}, $PM_{2.5}$ and $PM_{1.0}$. Grimm 31-Channel Portable Aerosol Spectrometer model No. 1.109 was selected for monitoring the mass concentration of PM_{10}, $PM_{2.5}$, $PM_{1.0}$ at a flow rate of 1.2 Liter/min ± 5% constant with controller for continuous measurement during the sampling period (www.GRIMM-aerosol.com). Sampling of PM was done by alternatively positioning the Grimm at different distances of each sampling site (due to lack of multiple samplers). A variation of 4% to 12% was observed in the mass concentration of PM when sampled continuously for one successive week at all sampling locations. The measurements of gases were also conducted by handy sampler APM 821(Envirotech, New Delhi), at a flow rate of 1.5 l/min. Sampling of gaseous pollutants was done simultaneously at different distances of each site by placing different samplers at each distance simultaneously. Quality assurance parameters, like collection efficiency and reproducibility, were measured. To check the efficiency of different handy samplers used in the study, three samplers used were placed together at one site for continually one week and the sampling of gaseous parameters was done constantly. Then the results of the three handy samplers were compared and the variation in the values of gaseous parameters found were as follows: 95–99% for SO_2, 93–97% for NO_2, 90–9% for NH_3 and 89–93% for O_3. Incomplete samples were discarded whenever there was power failure during sample collection.

Central Pollution Control Board (Central Laboratory test methods) was followed for analyzing gases using UV–visible spectrophotometer (Heλios-α, Thermospectronic, UK). Modified West-Geake method was applied for the determination of SO_2. The typical standard concentrations ranging between 0.5 and 2.0 ppm were used to plot a calibration curve. The accuracy of standard curve was 2%. The standards were run in triplicate. After every five samples the standard was run to check the peak response. If the deviation was > 2% recalibration was done. The analytical errors were nominal and varied within ± 10%. To determine collection efficiency, we sampled air into three impingers in a series and determined the concentration of SO_2 in each, separately. Maximum concentration

was found in the first impinger with a variation of 96–99% in the other impingers. Two parallel sets of measurements were carried out to determine the reproducibility with an identical absorbing solution. The differences of the parallel measurements under ambient conditions were < 4%. Concentration of NO_2 was determined by Saltzman's method as modified by Jacob and Hoccheiscer. The typical standard concentration ranged between 0.5 and 2.0 ppm. The same procedure, as in case of SO_2, was followed to obtain the calibration curve. The analytical errors were nominal and varied within ± 10%. To determine collection efficiency, the same technique as for SO_2 was used. Maximum concentration was found in the first impinger with a variation of 94–97% in the other two impingers. Two parallel sets of measurements were carried out to determine the reproducibility with an identical absorbing solution. The differences of the parallel measurements under ambient conditions were < 5%.

Air concentration of O_3 was determined by neutral KI buffer method. The typical standard concentration ranged between 0.5 and 2.0 ppm. The same procedure, as in case of SO_2, was followed to obtain the calibration curve. The analytical errors were nominal and varied within ± 10%. Impingers were shielded from direct sunlight by covering them with a black sheet to avoid photo-decomposition. Low temperature was maintained by putting the ice in the box, in which impingers were kept. To determine collection efficiency, the same technique as for SO_2, was used. Maximum concentration was found in the first impinger with a variation of 91–94% in the other two impingers. Two parallel sets of measurements were carried out to determine the reproducibility with an identical absorbing solution. The differences of the parallel measurements under ambient conditions were < 6%.

Air concentration of NH_3 was determined by Indo-phenol blue method. The same procedure, as in case of SO_2, was followed to obtain the calibration curve. The analytical errors were nominal and varied within ± 10%. To determine collection efficiency, the same technique as for SO_2, was used. Maximum concentration was found in the first impinger with a variation of 93–96% in the other two impingers. Two parallel sets of measurements were carried out to determine the reproducibility with an identical absorbing solution. The differences of the parallel measurements under ambient conditions were < 5%. Meteorology for the sampling period was taken from meteorological station (WM251, Envirotech make) installed close to the sampling sites at St. Johns College, Agra, at a height of 10 m from ground level for wind speed, wind direction, temperature and relative humidity.

2.3. Multivariate data analysis

Principal Component analysis (PCA) is a well-established tool for analyzing structure in multivariate data sets (Derwent et al., 1995). Beginning with a large number of correlated variables, it seeks to identify a small number of independent factors (i.e. principal factors) that can be used to explain the variance in the data. The number of extracted principal components corresponds to the number and the nature of the variables included. A varimax-rotated factor analysis was performed to identify the main sources influencing the concentration of the pollutants studied at the sampling sites. Due to the limited number of samples, single PCAs were performed for the whole year, instead of considering the single seasons separately. In this statistical method, a set of multiple intercorelated variables is replaced by a small number of independent variables (factors) by orthogonal transformations (rotations). This is achieved by diagnosing the correlation matrix of the variable, that is, by computing their eigenvalues and eigenvectors. Factor loadings obtained after the rotation called varimax rotation gives the correlation between the variables and the factors. Each variable was also evaluated for its Kaiser-Mayer-Olkin value (KMO) which was always > 0.7, which gives sampling adequacy, and data were included in the matrix only if it had eigenvalue > 1. The varimax procedure was adopted for rotation of the factor matrix to transfer the initial matrix into one that was easier to interpret. In the present study, SPSS (version 10.0) Computer software was used to perform factor analysis.

3. Results and discussion

3.1. Average concentrations of particulates and gases

The 24 h mean, median, standard deviation, maximum and minimum along with skewness for the gases NO_2, SO_2, NH_3 and particles PM_{10}, $PM_{2.5}$, $PM_{1.0}$, except O_3 for which 8 h mean was taken, are presented in Table 2. Large variation in ambient concentrations of pollutants was observed throughout the study at both the sites with skewness of 0.17 to 0.33 for NO_2, − 0.82 to 0.002 for SO_2, 0.41 to 0.56 for O_3, − 0.32 to 0.48 for NH_3, − 1.11 to − 0.04 for PM_{10}, − 0.17 to 1.14 for $PM_{2.5}$, 0.12 to 1.15 for $PM_{1.0}$, respectively (Table 1) [Skewness is the characterization of degree of asymmetry of a distribution around its mean. It is 3(mean - median)/Standard deviation. It ranges between − 3 to + 3 and skewness zero means a normal frequency curve] (Kulshrestha et al., 2009). The variation in pollutants concentration throughout the study is due to varying anthropogenic sources and meteorological conditions.

The mean values show negative association between distance and concentrations of NO_2, SO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$. This is supported by the fact that motor vehicle emissions are the greatest source for NO_2, PM and the third greatest source for SO_2 after heating and industry. In case of NO_2 and O_3 levels getting smaller as the distance from the highway increases. Since a high amount of traffic on the highway accounts for formation of high amount of NO and NO_2 which act as precursors for O_3 formation thus causes high level of O_3 near the highway (Volz and Kley, 1988). As the distance increases from the highway, we go towards the residential area with lesser amount of traffic. All these conditions cause low levels of NO and NO_2 which in turn accounts for low O_3 concentration far away from the highway (Hong and Cheng, 2008). The gradients of SO_2 also show a decreasing trend since SO_2 is usually emitted during the combustion of low quality fossil fuels and road traffic is one of the chief sources of fossil fuel pollution (Zou et al., 2007; Civan et al., 2015; Baldwin et al., 2015). For PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ a decreasing trend is obtained as we go farther away from the highway because particulate matter pollution is affected directly by tailpipe exhaust from motor vehicles, brake/tire

Table 2
Descriptive statistics of the pollutants measured near highway during sampling period.

Pollutants	A(0 m)	A(250 m)	A(500 m)	B(0 m)	B(250 m)	B(500 m)
$NO_2(\mu g/m^3)$						
N	47	46	46	43	45	40
Mean	33.24	22.88	18.47	27.86	18.86	14.87
Median	29.74	18.10	12.09	25.70	15.76	10.20
Max	98.04	94.04	95.52	98.72	89.97	80.51
Min	6.72	2.11	1.72	4.47	1.92	1.57
Stdev	20.28	19.15	18.93	17.63	15.50	14.87
Skewness	0.17	0.24	0.33	0.12	0.20	0.31
$SO_2(\mu g/m^3)$						
N	45	45	46	48	46	47
Mean	33.08	22.86	18.73	31.18	21.87	17.58
Median	38.15	25.38	18.72	35.47	23.25	19.88
Max	70.35	50.62	43.52	67.20	49.53	40.35
Min	6.12	2.85	2.07	6.31	3.11	2.42
Stdev	18.36	13.04	11.42	16.39	12.07	9.82
Skewness	− 0.82	− 0.57	0.002	− 0.78	− 0.34	− 0.70
$O_3(\mu g/m^3)$						
N	37	42	40	43	39	39
Mean	104.69	75.47	62.77	100.82	73.12	61.00
Median	92.68	62.63	50.07	86.81	58.90	48.01
Max	146.77	113.57	99.77	141.65	110.22	96.23
Min	70.82	46.38	36.55	69.25	47.58	37.01
Stdev	28.85	26.39	24.66	28.25	24.98	23.24
Skewness	0.41	0.48	0.51	0.49	0.56	0.55
$NH_3(\mu g/m^3)$						
N	43	40	45	46	44	41
Mean	9.47	8.26	7.85	8.77	6.34	10.04
Median	8.36	7.04	7.03	7.76	6.51	9.60
Max	22.62	23.87	20.17	25.31	26.41	30.98
Min	0.73	0.67	0.53	0.23	1.19	0.55
Stdev	6.17	5.80	5.34	6.07	3.51	7.13
Skewness	0.44	0.48	0.29	0.39	− 0.32	0.18
$PM_{10}(\mu g/m^3)$						
N	49	46	48	48	47	47
Mean	227.69	164.53	129.36	190.47	146.81	129.81
Median	231.05	199.75	151.35	192.20	159.10	135.99
Max	594.40	397.60	648.00	584.10	376.60	297.40
Min	41.30	18.80	14.70	19.60	17.60	16.30
Stdev	133.22	94.52	90.04	123.62	92.07	74.43
Skewness	− 0.07	− 1.11	− 0.73	− 0.04	− 0.40	− 0.24
$PM_{2.5}(\mu g/m^3)$						
N	49	46	48	48	47	47
Mean	144.57	98.94	76.10	122.56	87.83	77.23
Median	149.96	95.93	70.00	88.43	83.02	75.68
Max	345.89	282.60	174.50	310.84	201.67	171.70
Min	29.80	16.80	13.40	16.60	16.40	14.80
Stdev	89.93	67.66	55.01	89.15	60.59	51.85
Skewness	− 0.17	0.13	0.33	1.14	0.23	0.08
$PM_{1.0}(\mu g/m^3)$						
N	49	46	48	48	47	47
Mean	123.98	82.57	62.14	105.77	74.81	62.04
Median	111.44	59.20	59.27	81.03	61.40	60.14
Max	263.90	201.20	164.40	245.00	182.20	154.80
Min	23.80	14.90	6.20	10.30	10.50	9.90
Stdev	82.52	60.44	50.51	79.93	56.27	47.10
Skewness	0.45	1.15	0.17	0.92	0.71	0.12

N = Number of samples. *O_3 values are 8 h average.

wear and resuspended road dust (Wu et al., 2002).

We applied paired sample *t*-test for comparing mean-values of pollutants obtained at different distances from the highway. The p-value for NO_2, SO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$, $PM_{1.0}$ at A0-A250, B0-B250 and A250-A500, B250-B500 was < 0.01 which signifies 99% difference in their mean values. The above results confirmed that the average concentrations of all pollutants are decreasing with a significant difference in their values. Thus we can say that ambient concentrations are found higher close to highway and decline with

the distance from the highway. Further the maximum percent decrease in concentrations of NO_2, SO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$, $PM_{1.0}$ were 31.16, 30.89, 27.89, 12.77, 27.73, 31.56, 33.40 and 32.30, 29.85, 27.47, 27.70, 22.92, 28.33, 29.27 at 0–250 m distance of site A and site B respectively, over the total measured distance (0–500 m). It suggests that the major impact of traffic flow on the pollutants concentration lies within 250 m distance. Higher pollution on highway is attributed to diesel/petrol vehicular emissions, as the major source of air pollution.

The average concentrations were compared with national ambient air quality standards (NAAQS, 2009) and world health organization (WHO, Air quality Guidelines, 2006). The average concentration of PM_{10} was found to be 4 to 2 times higher and $PM_{2.5}$ was 3 to 2 times from 0 m to 500 m distance from the highway on comparing with standards prescribed by central pollution control board (CPCB), 2001. Levels of NO_2, O_3 and SO_2 were within permissible limits. The average concentration of PM_{10} was found to be 11 to 6 times higher and $PM_{2.5}$ was 14 to 7 times higher from 0 m to 500 m distance from the highway on comparing with WHO standards. These high levels of pollutants found near the highway account for respiratory problems found frequently in the residents living in close vicinity.

3.2. Monthly and seasonal variations

To see the seasonal effect, the seasonal average concentrations were shown in three seasons for each of the measured pollutants separately at 0 m, 250 m and 500 m from the highway (Fig. 2). The seasonal average concentrations were highest in winter followed by summer and lowest in Monsoon. The seasonal average concentrations of NO_2, SO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ shows a clear decline with distance from the NH-2 at both the sites. The highest value of NO_2 was found in December and the lowest in July. The highest value of SO_2 was found in January and lowest in July. The highest value of NH_3 was found in December and the lowest in June. The highest value of O_3 was found in November and the lowest in August. This pattern is similar to the results of the previous studies (Lee et al., 1999; Jo and Park, 2005). Although the wind speed varied little between seasons, other meteorological parameters, such as temperature and humidity, pointed to poorer mixing during winter (Derwent et al., 1995; Pecorari et al., 2013) resulting in higher concentrations of pollutants. More heating fuels are typically consumed during winter, thereby further elevating the ambient air levels of pollutants. Moreover, higher tailpipe emissions from motor vehicles are associated with cold ambient temperature, causing lower combustion efficiency of motor vehicle fuels in winter resulting in higher pollutants concentration (Bruetsch, 1981; Pecorari et al., 2013), thus accounting for higher levels of pollutants obtained in winter. During summer, the prevailing winds that are due to thermal circulations are stronger and the mixing height is deeper. Wind turbulence with large mixing heights results in proper dilution and dispersion of gases during summer (Mantis et al., 2005), thus accounts for moderate concentrations obtained during summer.

Higher particulate matter concentrations (both in coarse and fine sizes) were found mostly during winter. This might be due to low wind speed and high humidity during the season in comparison to other seasons. So the removal of aerosol particles by wet scavenging is reduced. Summers are marked by moderate particle levels due to high temperature and moderate wind speed results in greater diffusion of the particles. Minimum aerosol concentrations were found during monsoon because of wet deposition and washout of particles from the atmosphere. Similar seasonal variations were found by previous studies conducted in Kolkata (Karar and Gupta, 2006) and Mumbai (Gupta et al., 2004). The annual distribution of the monthly average concentrations for coarse particles (PM_{10}) is bimodal, the highest value being in April (425.50 $\mu g/m^3$ for PM_{10}). The second highest value of PM_{10} is in December with 376.77 $\mu g/m^3$ respectively. Their minimum values are in July with 55.70 $\mu g/m^3$. However, the annual distribution of PM concentrations for fine particles ($PM_{2.5}$ and $PM_{1.0}$) is unimodal, the highest value being in December (286.67 $\mu g/m^3$ and 255.83 $\mu g/m^3$ for $PM_{2.5}$ and $PM_{1.0}$ respectively). The lowest monthly average concentrations occur in June and are 40.49 $\mu g/m^3$ and 28.08 $\mu g/m^3$ for $PM_{2.5}$ and $PM_{1.0}$ respectively. The difference in the behavior between coarse and fine particles indicates that dust events in summer contributed more to coarse than to fine particles (Wang et al., 2009).

In order to further investigate the effect of seasonality on ambient air quality, winter to summer and winter to Monsoon, average ratios were calculated as shown in Table 3. It was observed that the concentrations values of NO_2, SO_2, O_3, NH_3, TSPM, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ in winters were 2.31 to 1.02 times higher than that in summers, respectively at site A (0 m), 3.22 to 1.20 times higher than that in summers, respectively at site A (250 m), 3.00 to 1.20 times higher, respectively at A (500 m), 3.33 to 1.35 times higher respectively at site B (0 m), 3.63 to 1.39 times higher respectively at B (250 m) and 3.45 to 1.33 times higher respectively at site B (500 m).

On comparing the seasonal average concentrations of winter to monsoon, it was observed that the concentration values of NO_2, SO_2, O_3, NH_3, TSPM, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ in winters were 5.22 to 1.60 times higher than that in the monsoon, respectively at site A (0 m), 5.45 to 1.31 times higher respectively at A (250 m), 7.22 to 1.40 times higher respectively at A (500 m), 4.46 to 1.78 times higher respectively at B (0 m), 5.32 to 1.61 times higher respectively at B (250 m) and 6.02 to 1.88 times higher respectively at B (500 m). It is apparent that there is a general pattern of increasing levels from monsoon to summer and from summer to winter and also on increasing distance from the highway.

The variation of monthly average concentrations with the meteorological parameters is shown in Fig. 3. From Fig. 3, it can be observed that pollutants levels were decreased on increasing temperature and wind speed, whereas not very good relationship was found between Pollutants level and humidity. In other words, the concentrations of pollutants were inversely proportional to temperature and wind speed (Jamriska et al., 2008). During the winter months (i.e. October to February), the concentrations were maximum due to low temperature, low wind speed and high humidity, then the concentrations were gradually decreased with increase in temperature and wind speed and decrease in humidity causing moderate concentrations during the summer months (March to June) and the minimum concentrations were found in monsoon (July to September) because of high temperature and high

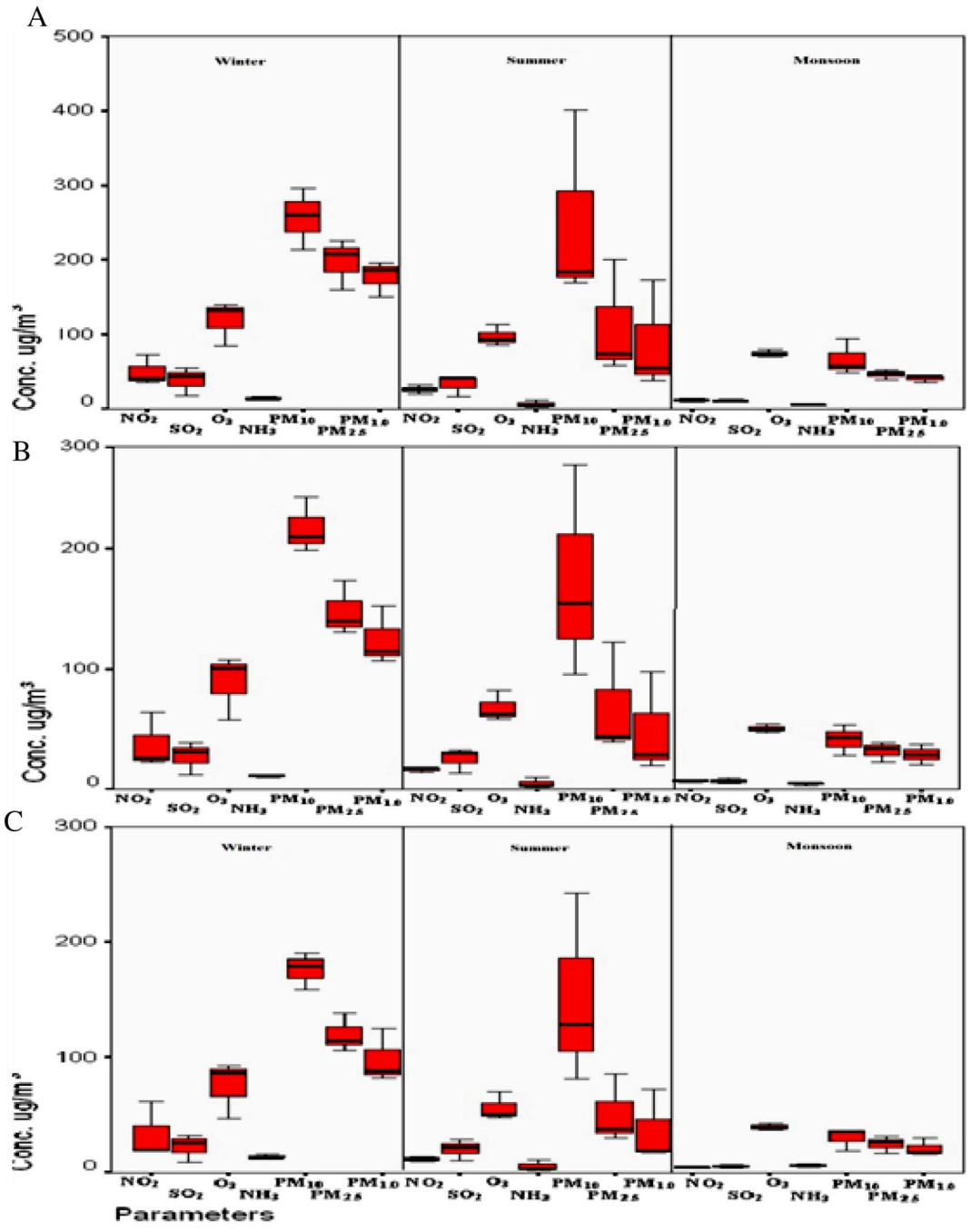

Fig. 2. A, B, C. Average Concentration of pollutants in three different seasons at 0 m, 250 m & 500 m distance.

humidity (Jo and Park, 2005). Almost similar pattern was observed at 0 m, 250 m and 500 m distances of both the sites (A and B).

3.3. Diurnal variation

The diurnal variation gives a clear picture of pollutants status near highway as shown in Fig. 4. The diurnal NO_2 averages exhibited a gradual increase in the pollution levels commencing around 6.00–7.00 h in the morning and culminating at approximately 9.00–11.00 h. The concentration then proceeded to a shallow trough before increasing once more to give a second peak concentration round 21.00–22.00 h. Thereafter the concentration decreased steadily as shown in Fig. 4A. This diurnal NO_2 trend is similar to the diurnal trend found in the Korean metropolitan city (Jo and Park, 2005) and at Azusa in California's south coast air basin (Fujita et al., 2003). Night time concentrations were found higher than daytime concentrations for NO_2. The night time increase

Table 3
Ratio of seasonal average concentrations of pollutants near highway.

Pollutants	Winter/summer ratio						Winter/monsoon ratio					
	A1	A2	A3	B1	B2	B3	A1	A2	A3	B1	B2	B3
NO_2	2.02	2.3	2.9	2.04	2.51	3.15	3.94	5.07	7.73	3.71	4.46	5.41
SO_2	1.26	1.25	1.38	1.23	1.18	1.16	3.98	4.52	5.08	3.63	4.06	4.16
O_3	1.78	1.96	2.00	1.82	1.97	1.90	3.07	3.38	3.25	3.25	3.25	3.17
NH_3	2.90	3.42	3.36	3.23	2.85	2.94	2.43	2.78	2.76	2.41	2.60	2.21
PM_{10}	1.21	1.47	1.44	1.55	1.58	1.50	3.09	5.71	8.60	4.65	5.35	4.95
$PM_{2.5}$	2.05	2.50	2.36	4.06	2.90	2.60	4.97	5.56	6.61	4.46	4.42	4.21
$PM_{1.0}$	2.31	3.22	3.00	3.33	3.63	3.45	5.23	5.45	6.17	4.21	3.63	4.56

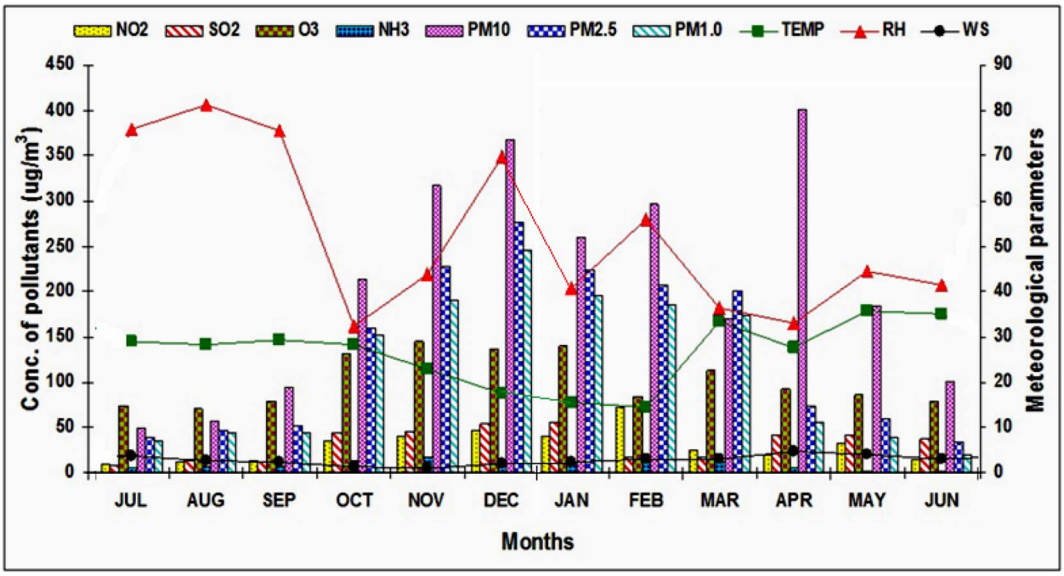

Fig. 3. Effect of meteorological parameters on the roadside average concentrations of pollutants from July 2009 to June 2010.

in NO_2 might be due to reduced mixing height (Chou et al., 2007). This result is also consistent with a previous roadside study conducted in Taiwan (Hong and Cheng, 2008).

Similar to NO_2, the diurnal trends of SO_2 were well-defined and exhibited a gradual increase in the pollution levels commencing around 7.00 h and culminating at approximately 11.00 h and then preceded to a shallow trough before increasing once more to give a second peak concentration around 20.00–22.00 h. Thereafter the concentration remained almost steady. This diurnal SO_2 trend is similar to the one found in the Korean metropolitan city (Jo and Park, 2005). Fig. 4.B shows the peak SO_2 levels coinciding with the traffic rush hours, the same as occur with NO_2.

The day time pattern for O_3 was different from those for NO_2 and SO_2. The ozone concentration starts increasing gradually after sunrise, attains the maximum value at noon and then declines. The rate of increase in O_3 concentration during morning is fast whereas in the evening it decreases rather slowly. This diurnal O_3 trend is similar to the one found in several other cities (Fujita et al., 2003). During the daytime, peak O_3 concentration coincide with flux of solar radiation indicating its photochemical production. Low levels of O_3 during early morning time could also be a result of its deposition and surface chemical reactions (Saini et al., 2008).

The diurnal NH_3 averages follows almost the similar pattern as for NO_2 with first peak in the morning 6.00–7.00 h then a shallow trough, before increasing once again to give a second peak in the evening around 21.00 h. The night time concentrations were found higher than the day time NH_3 concentrations. This can be attributed to the existence of stable atmosphere during night time (Cadle et al., 1982; Walker et al., 2004).

The diurnal pattern of the three kinds of particles (PM_{10}, $PM_{2.5}$ and $PM_{1.0}$) shows a bimodal distribution for the whole year. The first peak is observed before noon (9.00–10.00) and the secondary peak is observed before midnight (21.00–22.00). In general, the highest peaks of PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ occur at about the same time. However, there are obvious differences in the occurrence time of minimum concentrations among different kinds of PM. Low concentrations of all particles happen either early morning or late afternoon. The mass of a fine particle is relatively small, so it can suspend in the air for a long time. Thus the concentrations of fine particles are mainly affected by atmospheric diffusion conditions. In the afternoon, the dissipation of the boundary layer inversion and the development of a mixing layer are helpful for fine particles to diffuse, which results in the concentration minimum before dark. Compared with fine particles, coarse particles are heavier and cannot be suspended easily in the air for a long time. Thus, their

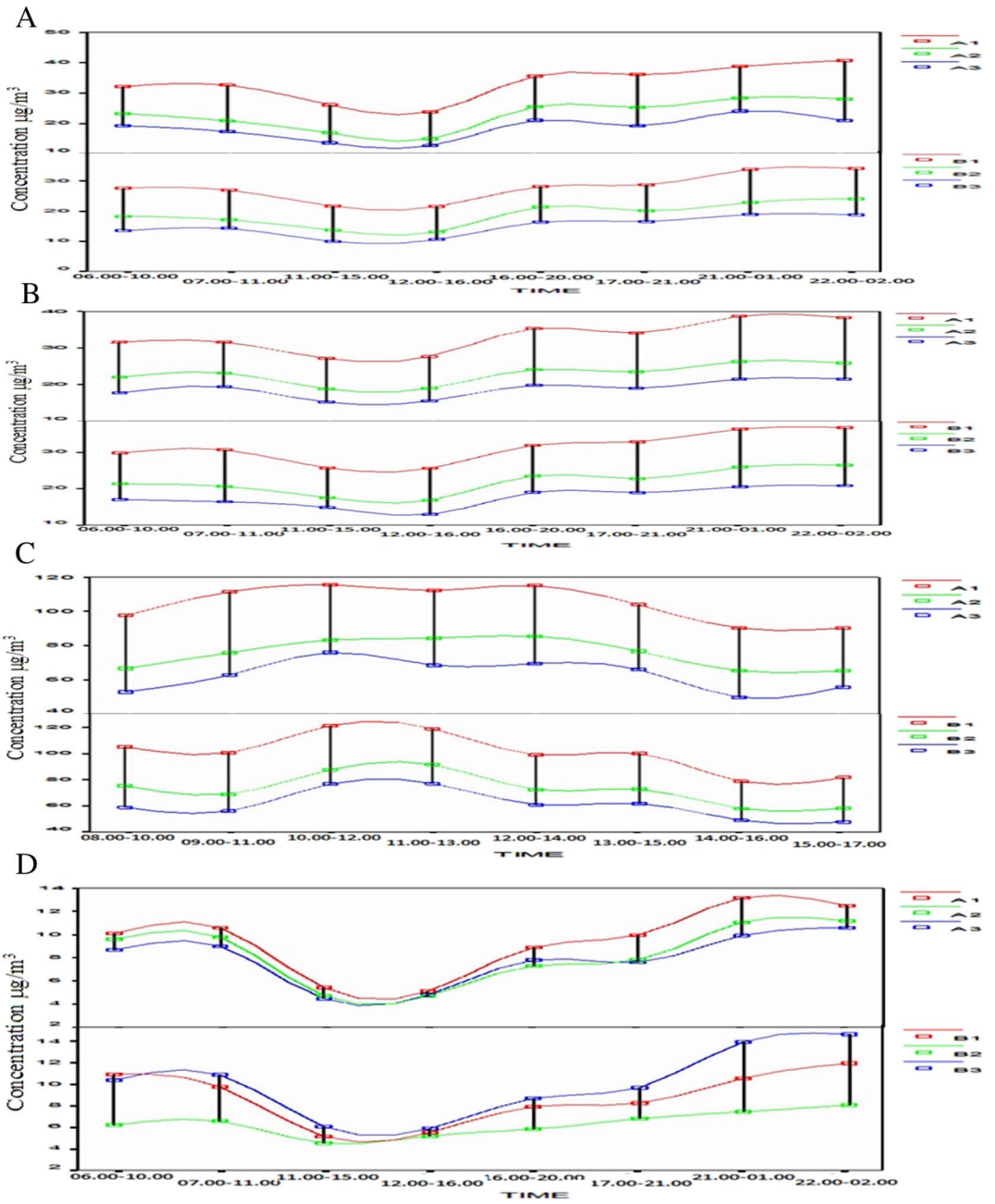

Fig. 4. A, B, C, D. Diurnal variation of NO_2, SO_2, $O_3$and NH_3at three different distances of the two sites A and B. E, F, G. Diurnal variation of PM_{10}, $PM_{2.5}$ & $PM_{1.0}$ at three different distances of the two sites A and B.

concentrations mainly depend on the variation of local emission. In general, the amount of local emission after midnight reaches the minimum of the day. On the other hand, re-suspension generated by traffic and other human activities also drops to the lowest point after midnight. As a result, the concentration of coarse particles reaches the minimum after midnight. This diurnal PM trend is similar to the one found in the China (Wang et al., 2009).

3.4. Correlation analysis

Statistical correlation among gaseous pollutants, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ at all the three distances of the two sites are presented in Tables 4.1, 4.2 and 4.3. The purpose of this study was to investigate the long-term characteristics of the atmospheric pollutant concentrations up to 500 m distance from the highway. Most of the variables were significantly correlated at 0.01 levels and 0.05 levels. NO_2 has strong to moderate association along with increasing distance from the highway, with PM_{10} (R ranges 0.66–0.49),

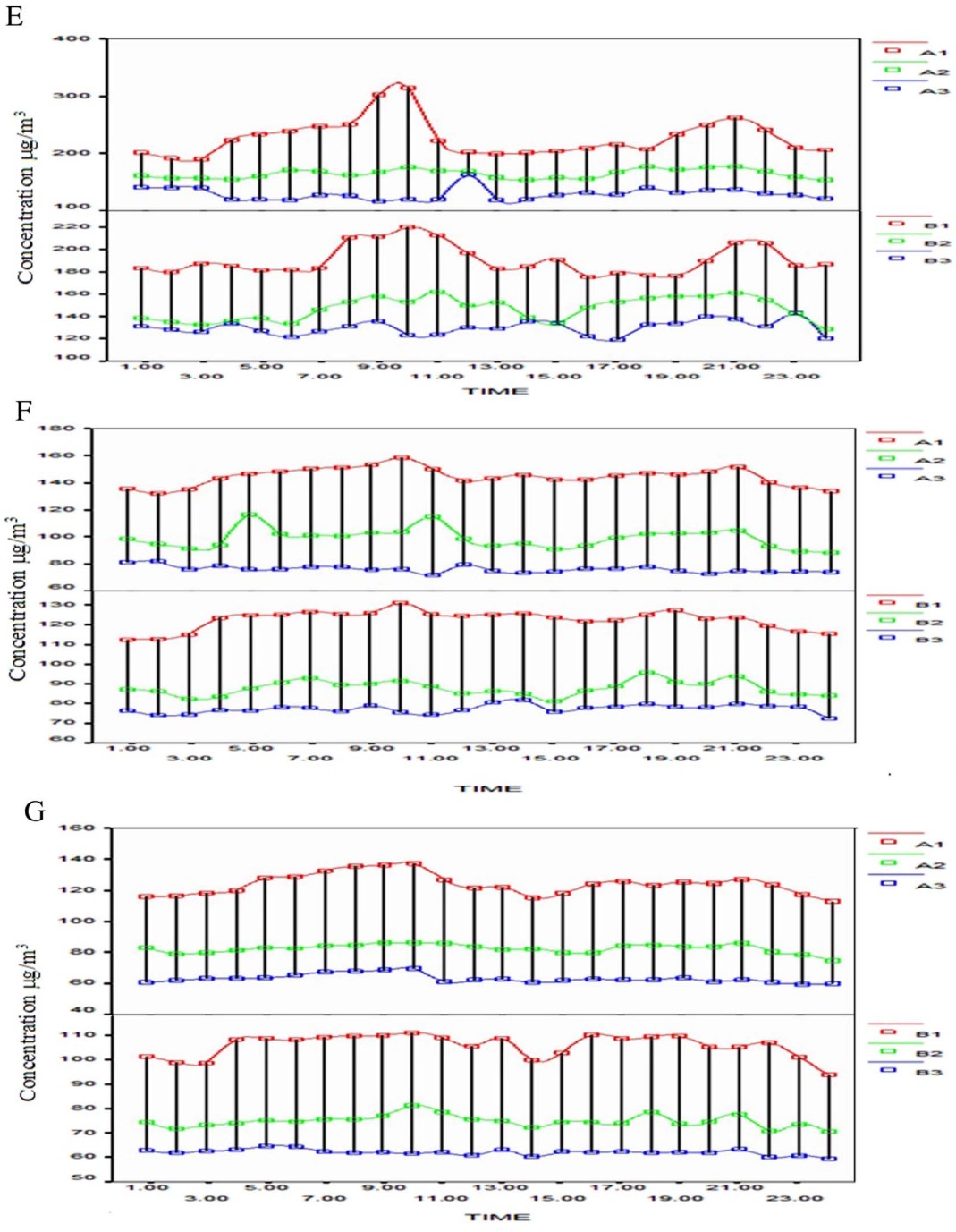

Fig. 4. (*continued*)

PM2.5 (R ranges 0.81–0.46), $PM_{1.0}$ (R ranges 0.80–0.47), NH_3 (R ranges 0.78–0.66). PM_{10} shows significant correlations with $PM_{2.5}$ (R ranges 0.70–0.60), $PM_{1.0}$ (R ranges 0.66–0.59), and SO_2 (R ranges 0.70–0.63). $PM_{2.5}$ shows a very high positive correlation at all of the sites with $PM_{1.0}$ (R ranges 0.99–0.98). The significant correlations indicate that the emission sources are to some extent similar i.e. vehicular exhaust generated by the traffic density affects pollutants in some or other way. For some of the pollutants (like NO_2, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$), it acts as a direct source of emission, while for other pollutants (like particulates, O_3 and SO_2) it affects their source of generation indirectly. It suggested that the vehicular emission, solid waste incineration, biomass burning and re-suspended road dust are the main probable sources near highway.

Table 4.1
Bivariate Pearson Correlation and sigma value (2-tailed) for site A and B at 0 m distance from highway.

	Correlations	A1						
		NO_2	SO_2	O_3	NH_3	PM_{10}	$PM_{2.5}$	$PM_{1.0}$
B1	NO_2	1.000	0.370	0.517	0.784**	0.648**	0.815**	0.802**
			0.236	0.085	0.003	0.023	0.001	0.002
	SO_2	0.237	1.000	0.745**	0.214	0.687*	0.513	0.508
		0.285		0.005	0.505	0.014	0.088	0.091
	O_3	0.398	0.717**	1.000	0.698	0.628*	0.847**	0.861**
		0.200	0.009		0.012	0.029	0.001	0.000
	NH_3	0.747**	0.163	0.643*	1.000	0.440	0.866**	0.885**
		0.005	0.613	0.024		0.153	0.000	0.000
	PM_{10}	0.634*	0.635*	0.592*	0.529	1.000	0.721**	0.679*
		0.027	0.026	0.043	0.077		0.008	0.015
	$PM_{2.5}$	0.699*	0.430	0.823**	0.866**	0.608*	1.000	0.996**
		0.011	0.163	0.001	0.000	0.036		0.000
	$PM_{1.0}$	0.714**	0.371	0.785**	0.884**	0.592*	0.995**	1.000
		0.009	0.235	0.003	0.000	0.043	0.000	

** Correlation is significant at the 0.01 level (2-tailed).
* Correlation is significant at the 0.05 level (2-tailed).

Table 4.2
Bivariate Pearson Correlation and sigma value (2-tailed) for site A and B at 250 m distance from highway.

	Correlations	A2						
		NO_2	SO_2	O_3	NH_3	PM_{10}	$PM_{2.5}$	$PM_{1.0}$
B2	NO_2	1.000	0.211	0.321	0.687*	0.668*	0.673*	0.653*
			0.509	0.308	0.013	0.017	0.016	0.021
	SO_2	0.218	1.000	0.749**	0.290	0.555	0.616*	0.576
		0.497		0.005	0.360	0.061	0.033	0.050
	O_3	0.313	0.689**	1.000	0.713**	0.778**	0.899**	0.894**
		0.321	0.013		0.009	0.003	0.000	0.000
	NH_3	0.645*	0.334	0.833**	1.000	0.740**	0.855**	0.848**
		0.023	0.289	0.001		0.006	0.000	0.000
	PM_{10}	0.691*	0.647*	0.598*	0.627*	1.000	0.883*	0.824*
		0.013	0.023	0.040	0.029		0.000	0.001
	$PM_{2.5}$	0.669*	0.448	0.859**	0.969**	0.677*	1.000	0.980**
		0.017	0.144	0.000	0.000	0.016		0.000
	$PM_{1.0}$	0.617*	0.442	0.849**	0.933**	0.647*	0.985**	1.000
		0.033	0.150	0.000	0.000	0.023	0.000	

** Correlation is significant at the 0.01 level (2-tailed).
* Correlation is significant at the 0.05 level (2-tailed).

3.5. Factor analysis

Results obtained by varimax-rotated factor analysis are given in Table 5 and the relationships among the seven pollutants (i.e. NO_2, SO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$) are readily seen. The factor analysis of the pollutants measured at 0 m distance from the highway reveals three factors with eigenvalue > 1 and contributes 82.71% of the total variance. The first factor contributed 46.50% of the total variance, which contains pollutants; NO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$, $PM_{1.0}$. It points at vehicular emission along with re-suspension of road dust as the main probable source (Beckerman et al., 2008; Hong and Cheng, 2008; Squizzato et al., 2012; Wu et al., 2014; Olayinka et al., 2015; Petracchini et al., 2016;). The second factor contributes to 21.48% of the total variance, explaining load of NO_2 and NH_3 was incineration since solid waste incineration or dumping can be commonly seen by the roadside mainly from nearby restaurants and hospitals (Walker et al., 2004; Beckerman et al., 2008; Hong and Cheng, 2008; Akagi et al., 2011; Petracchini et al., 2017). The third factor contributes 14.73% of the total variance, explaining load of SO_2 and O_3; this may be due to the biomass burning near highway and other anthropogenic activities, since the NO and CO emitted from burning, acts as precursor in ozone formation (Fujita et al., 2003; Saini et al., 2008; Saini et al., 2014; Civan et al., 2015; Baldwin et al., 2015).

The factor analysis of pollutants measured at 250 m distance from the highway reveals three factors with eigenvalue > 1 and contributes 84.14% of the total variance. The first factor, contributing to 48.67% of total variance, is mainly explained by the same variables of Factor 1 for 0 m distance. It can thus be attributed to the same emission source, i.e. road traffic emissions (Beckerman et al., 2008; Hong and Cheng, 2008; Squizzato et al., 2012; Wu et al., 2014; Olayinka et al., 2015; Petracchini et al., 2016). The second factor contributes to 20.69% of the total variance with large loading of NO_2 and NH_3 may be due to incineration because the blockage in open sewer lines due to accumulation of waste materials, within communities can be commonly seen (Walker et al., 2004;

Table 4.3
Bivariate Pearson Correlation and sigma value (2-tailed) for site A and B at 500 m distance from highway.

	Correlations	A3						
		NO_2	SO_2	O_3	NH_3	PM_{10}	$PM_{2.5}$	$PM_{1.0}$
B3	NO_2	1.000	0.128	0.264	0.663*	0.498	0.463	0.480
			0.691	0.407	0.019	0.099	0.130	0114
	SO_2	0.106	1.000	0.800**	0.350	0.599**	0.673*	0.650*
		0.742		0.002	0.265	0.039	0.016	0.022
	O_3	0.223	0.669**	1.000	0.739**	0.801**	0.942	0.926**
		0.487	0.017		0.006	0.002	0.000	0.000
	NH_3	0.696*	0.139	0.684*	1.000	0.715**	0.831**	0.812**
		0.012	0.666	0.014		0.009	0.001	0.001
	PM_{10}	0.478	0.704*	0.701*	0.508	1.000	0.892**	0.848**
		0.116	0.011	0.011	0.092		0.000	0.000
	$PM_{2.5}$	0.553	0.480	0.881**	0.886**	0.707*	1.000	0.988**
		0.062	0.114	0.000	0.000	0.010		0.000
	$PM_{1.0}$	0.469	0.462	0.881**	0.852**	0.661*	0.990**	1.000
		0.124	0.130	0.000	0.000	0.019	0.000	

** Correlation is significant at the 0.01 level (2-tailed).
* Correlation is significant at the 0.05 level (2-tailed).

Table 5
Results of factor analysis with varimax rotation at increasing distance from national highway.

Factors	0 m			250 m			500 m		
	1	2	3	1	2	3	1	2	3
NO_2	0.598	0.573	− 0.39	0.537	0.625	− 0.44	0.555	− 6.40E-03	0.745
SO_2	0.494	0.486	0.546	0.488	0.379	0.638	0.453	0.735	− 0.156
O_3	0.564	0.303	0.581	0.595	0.326	0.519	0.546	0.642	− 0.155
NH_3	0.615	0.498	− 0.477	0.657	0.528	2.80E-02	0.735	0.111	0.503
PM_{10}	0.662	− 0.501	8.30E-02	0.770	− 0.411	− 0.101	0.762	− 0.202	− 0.310
$PM_{2.5}$	0.887	− 0.420	− 6.59E-02	0.869	− 0.433	− 7.61E-02	0.898	− 0.322	− 0.217
$PM_{1.0}$	0.867	− 0.415	− 7.68E-02	0.865	− 0.423	− 7.79E-02	0.875	− 0.327	− 0.223
Eigen value	3.250	1.500	1.030	3.407	1.449	1.035	3.502	1.221	1.051
Total Variance	46.50%	21.48%	14.73%	48.67%	20.69%	14.78%	50.03%	17.45%	15.01%
Probable source	Vehicular emission	Incineration	Biomass burning	Vehicular emission	Incineration	Bio waste	Vehicular emission	Fuel burning	Incineration

Beckerman et al., 2008; Akagi et al., 2011; Petracchini et al., 2017). The third factor contributes 14.78% of the total variance; this may be due to accumulation of solid-waste, which includes open wastes, manholes and animal wastes explaining load of SO_2 and O_3 (Zou et al., 2007; Squizzato et al., 2012; Civan et al., 2015; Baldwin et al., 2015).

The factor analysis of pollutants measured at 500 m distance from the highway reveals three factors with eigenvalue > 1 and contributes 82.49% of the total variance. The first and most important factor that contains 50.03% of the total variance again consists of vehicular emission along with re-suspension of road dust as the main probable source, as it is responsible for the heavy loadings of NO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$, $PM_{1.0}$ (Beckerman et al., 2008; Hong and Cheng, 2008; Squizzato et al., 2012; Wu et al., 2014; Olayinka et al., 2015; Petracchini et al., 2016). The second factor contributes to 17.45% of the total variance, explaining load of SO_2 and O_3; fuel burning was the most probable source. Since it is a semi urban area and people used to burn kerosene, coal and cow dung cakes. Since fossil fuel emission is the direct source of SO_2 and indirect source of O_3 since the NO and CO emitted from fuel emission, acts as precursor in ozone formation (Zou et al., 2007; Saini et al., 2008; Saini et al., 2014; Civan et al., 2015; Baldwin et al., 2015). The third factor contributes 15.01% of the total variance this may be due to incineration and prominent dairy activities in that area, explaining the load of NO_2 and NH_3 (Walker et al., 2004; Beckerman et al., 2008; Akagi et al., 2011; Petracchini et al., 2017).

4. Conclusions

In the present study, the relationship between concentration of air pollutants and distance from a busy highway was observed. A negative association was found between distance and concentrations of NO_2, SO_2, O_3, NH_3, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$. The results of paired sample *t*-test confirmed that the average concentrations of all pollutants decrease with a significant difference in their values. The horizontal gradients at the highway suggested that pollutants concentrations were affected significantly by re-suspended road dust and tailpipe exhaust from motor vehicles. The pollutants level drop off rapidly in the first 250 m and then continue to decline slowly up to 500 m. The average concentrations were found highest in winter followed by summer and lowest in monsoon. The seasonal effect was further confirmed by seasonal average ratios of pollutants which show an increasing pattern from monsoon to

summer to winter. The diurnal pattern of pollutants shows a significant variation in concentration near highway (0 m) site with the traffic density at different times of the day since high concentrations were obtained with peak traffic hours. (first peak 7.00–10.00 in the morning because of large number of vehicles carrying school children and office employees and second peak at 20.00–22.00 in the evening due to office employees returning home and removal of No Entry restriction for heavy duty vehicles after 21.00 p.m.). This effect further diminished after 250 m distance and no proper trend was obtained. Degree of association of the pollutants with each other was assessed by bivariate pearson correlation. Most of the variables were significantly correlated at 0.01 levels and 0.05 levels. A varimax-rotated factor analysis was performed to identify the main sources influencing the concentration of the pollutants studied at the different sampling sites. The factor analysis of the pollutants measured at 0 m, 250 m and 500 m distance reveals three factors; the first and most prominent factor comprising vehicular emission along with re-suspended road dust up to 500 m distance from the highway. The other factors were variable and comprises of the pollutants measured at 250 m and 500 m distance from the highway follows almost the same pattern as the three factors identified were combustion activities, bio-waste accumulation and incineration explaining the load of the different pollutants in the residential area. These findings may have implications for studies related to health effects of traffic related pollutants.

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