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Temporal Variations of Ambient Air Pollutants Around Urea Fertilizer Plant in India During 2013-2015

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ABSTRACT

Monitoring of ambient air quality parameters (PM_{10}, $PM_{2.5}$ and NH_3) levels in the vicinity of a urea fertilizer plant located at Aonla, (U.P.) India, was carried out using the respirable dust sampler. The specific objectives were the determination of temporal variability of levels of ambient air pollutants and their relationship with the fertilizer plant emissions. Samples were collected for 3 consecutive years (2013-2015) from 5 different locations in the industrial area and the quantity of pollutants in the sample were experimentally analysed using the National Ambient Air Quality Standards recognized procedure, in an environmental management lab recognized by UP Pollution Control Board. The results showed a marked seasonal trend and temporal variability of pollutant levels in the study area that were higher in winter and lower in summer, while in rainy period levels were far less than other seasons. Annual mean PM_{10}, $PM_{2.5}$ and NH_3 concentrations were found to be 30.574 µg/m^3, 8.109 µg/m^3, 22.024 µg/m^3 for year 2013, 30.046 µg/m^3, 8.129 µg/m^3, 22.220 µg/m^3 for year 2014 and 30.324 µg/m^3, 8.361 µg/m^3, 23.254 µg/m^3 for year 2015 respectively. The concentration in the study region was jointly influenced by anthropogenic emission and regional natural processes, especially dust events and precipitation. The 24-hr values for PM_{10}, $PM_{2.5}$ and NH_3 in all the years from 2013 to 2015 were found to be lower than the prescribed limits of National Ambient Air Quality Standards, which prove the efficiency of the existing pollution control systems being implemented in the plant premises and the scope for new technology which could help in further lowering the emissions.

INTRODUCTION

The world is facing environmental crisis due to severe air pollution which is a result of rapid urbanization and industrialization, an unprecedented surge in automobile sector, and boom in population and economic expansion (Gupta et al. 2007). Deteriorating air quality is a major problem faced by millions of Indians, as most of the Indian cities are highly polluted with pollutant concentrations well above the recommended limits of WHO and US EPA (Gupta & Kumar 2006). Pollutant levels are monitored in ambient air quality networks because of their potential impact on human health, visibility and climate (Sharma & Pervez 2002). In India, major sources of air pollution include soil-derived aerosols, automobile emissions, industrial processes and coal burning (Kothai et al. 2009). Increased industrialization could be responsible for current elevated levels of pollutants than their natural occurrence in the environment (Leili et al. 2008). Fertilizer industry is one of the major emission sources of toxic metal-loaded ambient PM_{10}, $PM_{2.5}$, NH_3, NOx and SO_2, which arise chiefly from ammonia and urea production, as India is the largest manufacturer and highest consumer of fertilizers in the world (Sharma & Pervez 2002).

In the last decade, a review of the national air quality standards has been carried out in many countries, and the results of many studies showed a close relationship between pollutants and meteorological parameters, the most important is in the dispersion, transformation and removal of air pollutants from the ambient atmosphere. In order to develop an environmental forecasting tool, Slini et al. (2003) investigated the correlation of air pollution and meteorological data using neural networks, helping to understand the relationships between the atmospheric circulation, local meteorology and concentrations of troposphere air pollutants. The relationships between synoptic meteorology and air pollution have also been investigated for pollutants such as ozone (Krupa et al. 2003), SO_2 (Ocak & Turalioglu 2008), NO_2 (Perez & Trier 2001) and visibility (Sequeria & Lai 1998).

A number of previous studies on temporal and spatial variations of concentrations of air pollutants have been reported. Qu et al. (2010), studied spatial and temporal distribution of PM_{10} reconstructed from reported API records from 2000 to 2006 in 83 cities and found that PM_{10} concentrations peaked in the winter and lowest in the summer. Studies on spatial and temporal distribution of multiple criteria

pollutants have also been reported by Chai et al. (2014), for 6 criteria pollutants in 26 cities in China using data collected between August 2011 and February 2012. All pollutants except ozone (O_3) showed higher concentrations during winter months and lower concentrations in the summer. Carbon monoxide (CO), SO_2, $PM_{2.5}$ and PM_{10} were much higher in northern cities, but O_3 and NO_2 did not show significant differences between northern and southern cities. Ji et al. (2012) conducted a more detailed analysis of regional PM events in northern China during October and November 2009 using PM_{10}, SO_2 and NOx data collected at 24 sites and $PM_{2.5}$ data collected at 5 sites and concluded that light, wind, elevated temperature in surface inversion and low mixing height caused by low pressure systems were responsible for the weak dilution of primary pollutants and enhanced secondary PM formation. Although these studies provide valuable insights, none of them covers a full-year data and few of them include cities in less economically developed areas.

Gas-phase ammonia (NH_3) is an abundant alkaline gas that plays a significant role for aerosol formation in the atmosphere (Behera et al. 2013). Ambient NH_3 comes from biogenic sources as well as anthropogenic sources, including soil and vegetation, livestock operations, mobile exhaust and fertilizer application (Shen et al. 2011). Recent evidence suggests that industrial processes, human activities and vehicular emissions may have a greater impact on urban atmosphere, where ambient levels of NH_3 were found to be higher in more densely populated areas (Alebic-Juretic 2008). The investigation of NH_3 effects on urban air is of great importance for addressing environmental issues.

The critical level of NH_3 in ecosystems was set to be 8 $\mu g/m^3$ (or 0.011 ppmv) as an annual average (Reche et al. 2012). Huang et al. (2012) reported NH_3 concentration in an office as high as 6 ppmv, which definitely has impact on health. More importantly, NH_3 is one the important precursor of inorganic fine particles, once emitted to the atmosphere, NH_3 reacts with acidic gases such as nitrogen oxides (NOx) or sulphur oxides (SOx) to form secondary ammonium aerosols, typically in the fine particle matter size range of $PM_{2.5}$ (Ianniello et al. 2011). NH_3 plays a significant role in the neutralization of anthropogenic acidity in the atmosphere (Li et al. 2014), affects ultraviolet visibility, ambient acidity, eutrophication and biodiversity of ecosystems. The NH_3 emission control was not paid attention for a long time in our country, resulting in an increase in the formation of ammonium aerosols.

Thus, in the present study, concentrations of different pollutants (PM_{10}, $PM_{2.5}$ and NH_3) were measured during 2013-2015 at 5 locations around the fertilizer industry. Between January 2013 to December 2015, the temporal and spatial variations of $PM_{2.5}$, PM_{10} and NH_3 and the air quality attainment conditions were studied and the leading factor that exceeds the ambient air quality standard was determined. Inter-correlation of different pollutants was studied to provide a more comprehensive understanding of the current status of air pollution.

MATERIALS AND METHODS

Site selection: The present research was conducted in Aonla plant of Indian Farmers Fertiliser Cooperative Limited (IFFCO), the site is 28 km southwest of Bareilly on Bareilly Aonla Road in the State of Uttar Pradesh, India (Fig. 1). The site selection was based on importance of emission sources, sensitivity of receptors, predominant local activities and wind directions in the area (Gupta et al. 2007). The emission air samples were collected from 5 different locations in the industry area and the quantity of pollutants in the sample were experimentally analysed in the environmental management lab recognized by UP Pollution Control Board.

Fig. 1: Aerial location of IFFCO plant, Aonla, India (source: Google maps).

The data for 3 consecutive years (2013-2015) were collected and the emission trend was studied for these years along with the effect of environmental parameters on emission concentration and dispersion. The stations were so chosen that there can be adequate safety measures as well as reduced interference of the local public with the devices used for the experiment. The monitoring stations chosen were:

Station-1: North side of holding pond
Station-2: Laboratory
Station-3: South side of cooling tower II
Station-4: Raw water reservoir
Station-5: Residential colony

Sampling: For PM and NH_3, air samples were collected from January 2013 to December 2015, each PM sample was collected on a Whatman glass micro fibre filter of 20.3 cm × 25.4 cm size having a low resistance to airflow, a low affinity for moisture and a 98 percent collection efficiency for particles of 0.5 µ or large size (Sharma & Pervez 2002) using respirable dust sampler (model: APM 460NL, Envirotech, Delhi, India). Instantaneous airborne NH_3 was determined on a 24 h basis by using a monitoring instrument APM 411 TE designed as an attachment to operate with respirable dust samplers.

The sampling height was about 10 m from the ground level and operated 8 h round a clock (24 h) at an average flow rate of 1-1.2 m^3/min for PM and 1 L/min for NH_3 a day twice in a week. Sampler flow through the manometer was checked hourly, and the pump flow was calibrated once a month. Filters were conditioned in desiccators containing silica gel for 24 h before and after sampling. The daily averaged PM concentration was determined gravimetrically.

Sample analysis: The total air samples from 5 locations were analysed gravimetrically by pre-calibrated electronic balance of 10 µg precision to determine mass concentrations of PM using pre and post sampling filter weights taking into account the sampled air volume. The filters were equilibrated under controlled temperature (20-30°C) and relative humidity (40-50 percent) 48 h prior to weighing, in order to ensure consistent values of particulate mass (Sharma & Pervez 2002). Field and laboratory blank filters were also used to ensure accuracy of weights.

For NH_3, prescribed indophenols blue method was used. Filters were extracted in 25 mL distilled water maintaining the temperature at 25°C and pH by adding buffer solution (2 mL), then adding working phenol solution (5 mL) along with hypochlorite solution (2.5 mL) and standing for 30 min in darkness to develop colour and then measuring the absorbance of solution at 630 nm using UV spectrophotometer. A blank was also run to calculate the concentration of NH_3 in µg /m^3.

Statistical analysis: To evaluate the relationship between PM_{10}, $PM_{2.5}$ and NH_3, various multivariate techniques were used like factor analysis, a commonly used technique over last two decades, was basically a principal component analysis which explains maximum total variance of the data set and shows high correlation with one set of variables and little or no correlation with another set of variables.

The data were entered into STPR version-2 software for statistical analysis and two factorial analysis of variance (ANOVA) was performed. In order to compare the temporal variability of PM and NH_3 across five sites for three different years, coefficient of temporal variation was calculated for PM_{10}, $PM_{2.5}$ and NH_3 for each month, year as well as for whole period of sampling. The probability value of $P<0.05$ was set as the level for statistical significance. For each size fraction the overall mean of the five sites for each month of all 3 years and corresponding standard deviation were computed.

RESULTS AND DISCUSSION

Monitoring of 3 ambient air quality parameters (PM_{10}, $PM_{2.5}$ and NH_3) around a urea fertilizer plant in 12 months (January-December) of 3 years (2013, 2014 and 2015) was documented. Results were discussed on the basis of mean concentration values of PM_{10}, $PM_{2.5}$ and NH_3 air pollutants. The intent for this research was to provide data and give an overview of long term pollutant level in an industrial area. Table 1 gives the total and annual mean PM_{10}, $PM_{2.5}$ and NH_3 concentrations as well as the annual data completeness.

Results for PM_{10} for months (January-December) were found to be significant at 5% and 1% level of significance (0.892 and 1.176 respectively), for $PM_{2.5}$ (0.702 and 0.925 respectively) and for NH_3 (0.679 and 0.896 respectively). The mean concentration for PM_{10} was found to be highest in the month of December (39.037 µg/m^3), as the production of urea fertilizer was highest in December, and the lowering of temperature and wind speed in winter season resulted in higher concentration and low dispersion of particulate in the area, followed by the month of January (35.741 µg/m^3), where the high levels of PM_{10} can be reasonably attributed to the effects of reduced mixing height common in winter season (Yang et al. 2015), high concentration in November (35.481 µg/m^3) and October (32.222 µg/m^3), indicate that fireworks during the Diwali festival in India affected the ambient air quality adversely due to emission and accumulation of PM_{10} (Singh et al. 2010), which were found to be 2-4 times increase in concentration, followed by high levels in March (31.926 µg/m^3) and April (31.889 µg/m^3) due to high production of urea. The lesser concentration was found in the months of July and August (22.333 µg/m^3 and 22.704 µg/m^3 respec-

tively), consequent to maximal precipitation during this period which helps to lower the particulate level as being washed out by rainfall. A considerable amount of concentration was found in the months of February, May, June and September (28.259 μg/m^3, 27.815 μg/m^3, 29.852 μg/m^3 and 26.519 μg/m^3 respectively), owing to the high production in February and September, but the unusually high number of dust storm events in months of May and June substantially enhanced the particle levels. Sharma & Pervez (2002) also studied the spatial variability of ambient particulate matter around fertilizer plant and concluded that marked spatial and seasonal variability trend is seen in the study area with high PM_{10} level in winter season and lower in summer, while in rainy season levels were far less than other seasons.

The mean concentration for $PM_{2.5}$ was found to be highest in the month of December (10.629 μg/m^3), owing to the high pollutant levels of $PM_{2.5}$ often occurred in winter and on days with high haze, followed by the months of November (10.259 μg/m^3), January (10.000 μg/m^3), October (9.037 μg/m^3), characterized by strong amplitudes mostly related to changes in weather conditions, the concentration levels could rise due to accumulation of pollutants in the mixing layer. The lesser concentration was found in the months of July and August (6.222 μg/m^3 and 6.519 μg/m^3 respectively) owing to high amount of rainfall over the region which helps to settle down the particles on ground. A considerable amount of $PM_{2.5}$ concentration was found in the months of June, March, May, February, April and September (8.593 μg/m^3, 8.074 μg/m^3, 7.519 μg/m^3, 7.259 μg/m^3, 7.252 μg/m^3 and 7.037 μg/m^3 respectively), owing to the pollutants getting airborne from the ground surface. Their residence in the ambient atmosphere is controlled not only by the rate of source-emission, but also by wind speed, turbulence level, air temperature, and precipitation (Giri et al. 2008), and also by re-suspension of particulates owing to vehicular movement, as particles which have settled on ground surface get dislodged and get airborne by such frequent turbulences. Once getting airborne, the particulates get influenced by the prevailing climatic conditions, inferring that it has influence on weather and weather also having its own influence on pollutant concentration, distribution and translocation. A similar study was done by Liu et al. (2015), who studied the seasonal and diurnal variation in the particulate matter and concluded in 9-year observation period that PM_{10} and $PM_{2.5}$ showed high levels, with annual mean values of 138.5±92.9 and 72.3±54.4 μg/m^3, respectively. Slight decreasing trends of annual mean PM_{10} and $PM_{2.5}$ were observed in autumn, whereas lower values were found in the summer and spring months, respectively.

Gaseous ammonia (NH_3) is the most abundant alkaline gas in the atmosphere and a major component of total reactive nitrogen. The large sources of NH_3 emissions are industrial processes, vehicular emissions, agriculture, including animal husbandry and fertilizer applications. From the experimental work done, the mean concentration for NH_3 was found to be highest in the month of December (28.927 μg/m^3), followed by the months of January (27.989 μg/m^3), November (25.101 μg/m^3) and March (23.887 μg/m^3), due to the high production of ammonia during these months in the fertilizer plant along with the contribution of all forms of transport on the Bareilly-Aonla road on which the plant is situated, as during winter the low wind speed and temperature did not allow the dispersion of pollutants. The lesser concentration was found to be in the months of May, June and July (17.108 μg/m^3, 18.188 μg/m^3 and 19.977 μg/m^3 respectively), as the ammonia production was less during these months and the wind speed was high which allowed the far away dispersion of the pollutants. A considerable amount of concentration was found in the months of October, September, August, April and February (22.738 μg/m^3, 22.102 μg/m^3, 21.118 μg/m^3, 21.168 μg/m^3 and 21.694 μg/m^3 respectively), as there are other sources of emissions along with the production of ammonia which contribute to such high level of NH_3 pollutant like transport on Bareilly-Aonla road, residential colony situated along with the fertilizer plant, industrial processes (non-combustion) and product use, which cover production of chemicals used in plant, agriculture sector consisting of livestock manure management and application of manure and synthetic fertilizers on field, agricultural waste burning, as the fertilizer plant is surrounded by the agricultural fields where cultivation of crops is done for whole year and other waste handling in plant premises. The same results were also reported by Wang et al. (2016), who monitored the gas-phase ammonia (NH_3) and fine particle ($PM_{2.5}$) ammonium (NH_4^+) (collectively, NHx) between July 2013 and August 2014 in a busy traffic area of Nanjing, China. Results showed that $PM_{2.5}$ concentration was 66.7 μg/m^3 and NH_3 concentration was 6.66 μg / m^3, and concluded that traffic intensity in the region was partially related to the formation of $PM_{2.5}$ and NH_3, suggesting that traffic pollution may be an important source of NH_3 and $PM_{2.5}$.

The results for PM_{10} for 3 years (2013-2015) were not found to be significant at 5% and 1% level (0.446 and 0.588 respectively) and for $PM_{2.5}$ (0.351 and 0.462 respectively), but significant for NH_3 (0.339 and 0.448 respectively) (Table 1). The annual mean concentration of PM_{10} was found to be maximum in the year 2013 followed by 2015 and 2014 (30.574 μg/m^3, 30.324 μg/m^3, 30.046 μg/m^3 respectively), for $PM_{2.5}$ highest was found in year 2015, then in year 2014 and 2013 (8.361 μg/m^3, 8.129 μg/m^3 and 8.109 μg/m^3 respectively), and for NH_3 in year 2015, then in year 2014 and 2013

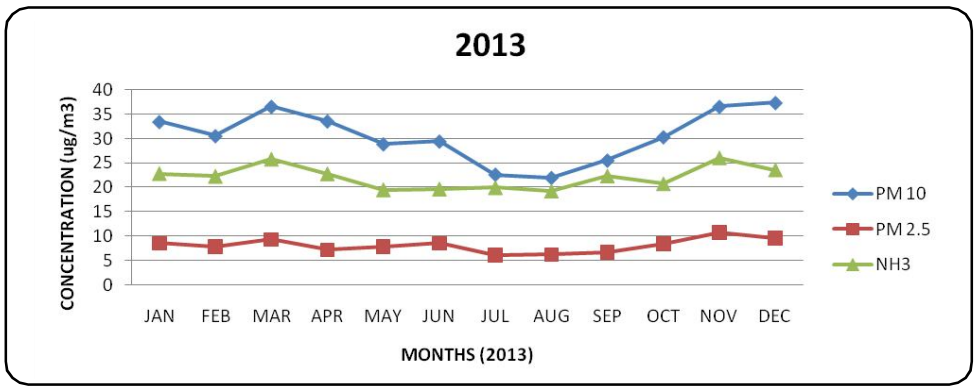

Fig. 2: Trend of monthly average of PM_{10}, $PM_{2.5}$ and NH_3 in year 2013.

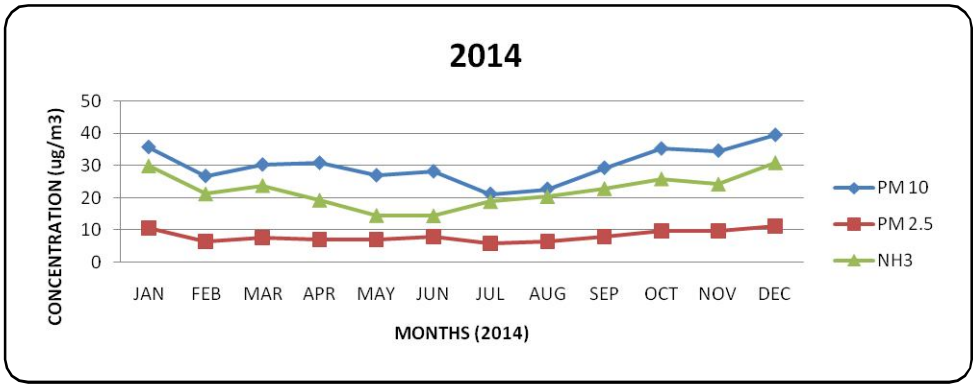

Fig. 3: Trend of monthly average of PM_{10}, $PM_{2.5}$ and NH_3 in year 2014.

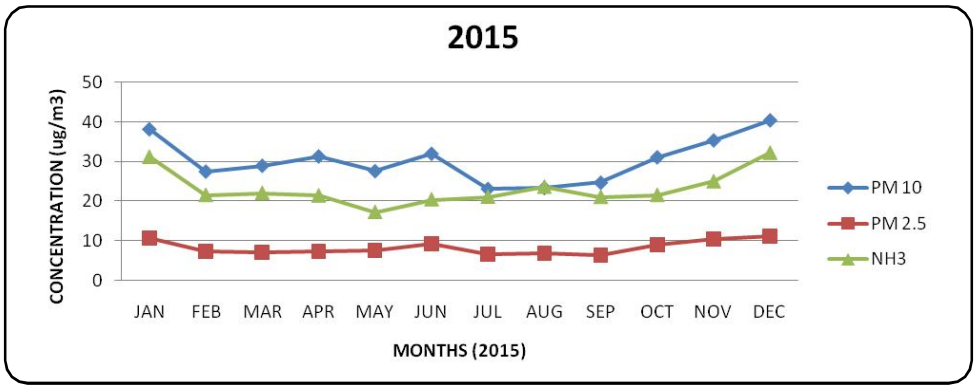

Fig. 4: Trend of monthly average of PM_{10}, $PM_{2.5}$ and NH_3 in year 2015.

(23.254 µg/m^3, 22.220 µg/m^3 and 22.024 µg/m^3 respectively), owing to higher concentrations during winter could be attributed by combined effect of the elevated levels of fossil fuel in plant production, biomass and coal burning in the residential colony and surrounding area alongside of plant and the prevailing atmospheric conditions such as low temperature and mixing height, low or calm wind speed, and temperature inversions which favour more accumulation, limit the dilutions, and dispersions of pollutants in atmosphere. Similar trend in pollutant levels has also been identified and discussed in previous studies (Shi et al. 2003, Sun et al. 2004, Gupta & Kumar 2006, Chatterjee et al. 2007, Wang et al. 2008, Kothai et al. 2009, Huang et al. 2010).

For interaction between year and month, all results were found to be significant at 5% and 1% level (1.546 and 2.036 respectively) for PM_{10}, for $PM_{2.5}$ (1.216 and 1.601 respec-

Table 1: The total and annual mean PM_{10}, $PM_{2.5}$ and NH_3 concentrations (μg/m³) as well as the annual data completeness.

	PM_{10}				$PM_{2.5}$				NH_3			
Month/ Year	2013	2014	2015	Mean	2013	2014	2015	Mean	2013	2014	2015	Mean
Jan	33.444	35.667	38.111	35.741	8.556	10.667	10.778	10.000	22.770	29.931	31.264	27.989
Feb	30.556	26.667	27.556	28.259	7.889	6.556	7.333	7.259	22.231	21.260	21.590	21.694
Mar	36.556	30.222	29.000	31.926	9.333	7.667	7.222	8.074	25.703	23.840	22.119	23.887
Apr	33.556	30.778	31.333	31.889	7.200	7.111	7.444	7.252	22.736	19.303	21.464	21.168
May	28.889	26.889	27.667	27.815	7.889	7.000	7.667	7.519	19.504	14.544	17.276	17.108
Jun	29.444	28.111	32.000	29.852	8.556	8.000	9.222	8.593	19.650	14.434	20.479	18.188
Jul	22.667	21.111	23.222	22.333	6.111	5.889	6.667	6.222	19.923	18.838	21.171	19.977
Aug	22.000	22.667	23.444	22.704	6.222	6.445	6.889	6.519	19.222	20.454	23.677	21.118
Sept	25.556	29.222	24.778	26.519	6.778	7.889	6.444	7.037	22.318	22.893	21.096	22.102
Oct	30.333	35.222	31.111	32.222	8.444	9.667	9.000	9.037	20.709	25.908	21.598	22.738
Nov	36.556	34.556	35.333	35.481	10.778	9.556	10.444	10.259	25.978	24.294	25.031	25.101
Dec	37.333	39.444	40.333	39.037	9.556	11.111	11.222	10.629	23.547	30.950	32.283	28.927
Mean	30.574	30.046	30.324	30.315	8.109	8.129	8.361	8.200	22.024	22.220	23.254	22.499
	Year	Month	Year*Month		Year	Month	Year*Month		Year	Month	Year*Month	
SEM	0.160	0.321	0.555		0.126	0.252	0.437		0.122	0.244	0.423	
CD At 1%	0.588	1.176	2.036		0.462	0.925	1.601		0.448	0.896	1.551	
CD At 5%	0.446	0.892	1.546		0.351	0.702	1.216		0.339	0.679	1.178	
CV	5.495				15.977				5.642			

tively) and for NH_3 (1.178 and 1.551 respectively) (Table 1). For the year 2013, the maximum values for PM_{10}, $PM_{2.5}$ and NH_3 were recorded in the months of December (37.333 μg/m³), November (10.778 μg/m³) and November (25.978 μg/m³) respectively, followed by the November (36.556 μg/m³) and March (36.556 μg/m³) for PM_{10}, December (9.556 μg/m³) and March (9.333 μg/m³) for $PM_{2.5}$, March (25.703 μg/m3) and December (23.547 μg/m³) for NH_3 respectively. The minimum values were recorded in the months of July (22.667 μg/m³) and August (22.000 μg/m³) for PM_{10}, June (6.111 μg/m³) and July (6.222 μg/m³) for $PM_{2.5}$ and August (19.222 μg/m³) and May (19.504 μg/m³) for NH_3 (Fig. 2). Many studies indicated that particulate matter concentration in ambient air is affected by temperature, wind speed, wind direction, solar radiation, relative humidity and rainfall (Monn 2001). The similar results were also reported by Singh & Sharma (2012) who concluded that the annual average concentration of pollutants showed a significant variation, found higher in winter >summer >monsoon.

For the year 2014, the maximum values for PM_{10}, $PM_{2.5}$ and NH_3 were recorded in the month of December (39.444 μg/m³, 11.111 μg/m³ and 30.950 μg/m³ respectively), followed by January (35.667 μg/m³) and October (35.222 μg/m³) for PM_{10}, January (10.667 μg/m³) and October (9.667 μg/m³) for $PM_{2.5}$, January (29.931 μg/m³) and October (25.908 μg/m³) for NH_3 respectively. The minimum values were recorded in the months of July (21.111 μg/m³) and August (22.667 μg/m³) for PM_{10}, July (5.889 μg/m³) and August (6.445 μg/m³) for $PM_{2.5}$ and May (14.544 μg/m³) and June (14.434 μg/m³) for NH_3 (Fig. 3). Wang et al. (2014), examined the spatial and temporal variations of $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2 and 8h O_3 in 31 capital cities in China between March 2013 and February 2014 using hourly data released by the Ministry of Environmental Protection (MEP) of China, and found that the annual mean concentrations of $PM_{2.5}$ and PM_{10} exceeded the Chinese Ambient Air Quality Standards (CAAQS); the observed $PM_{2.5}$, PM_{10}, CO and SO_2 concentrations were higher in cities located in the north region than those in the west and the south-east regions and were highest in the winter, then during the spring.

For the year 2015, the maximum values for PM_{10}, $PM_{2.5}$ and NH_3 were recorded in the months of December (40.333, 11.222 and 32.283 μg/m³ respectively), followed by January (38.111 μg/m³) and November (35.333 μg/m³) for PM_{10}, January (10.778 μg/m³) and November (10.444 μg/m³) for $PM_{2.5}$, January (31.264 μg/m³) and November (25.031 μg/m³) for NH_3 respectively. While the minimum values were recorded in the months of July (23.222 μg/m³) and August (23.444 μg/m³) for PM_{10}, September (6.444 μg/m³) and July (6.667 μg/m³) for $PM_{2.5}$, May (17.276 μg/m³) and June (20.479 μg/m³) for NH_3 (Fig. 4). In agreement to the results, Khamdan et al. (2009) used the non-parametric Kruskal–Wallis (KW) and Mann Whitney (MW) test to show significant spatial variations and interactions of spatial-temporal among five mobile monitoring stations for 11 air pollutants and concluded that emissions from automobile exhaust, in-

dustrial and developmental projects are responsible for the spatial air pollution, and temperature is the controlling factor for the seasonal variations.

Among many weather parameters, temperature and wind direction were two main factors influencing pollutant load, as a study in Netherlands has reported (Van der Wal et al. 2000). Another report from Hongkong relates the influence of monsoon and rain (Chang et al. 2000). In the south of Spain, a study related the influence of wind direction on PM concentrations, inferring that the sources for PM pollution were external and spread over the area (Gallero et al. 2006).

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