



Assessment of the Annual and Seasonal Trends of Gaseous & Particulate Pollutants around a Fertilizer Plant in India: A Climatic Perspective

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Authors' contributions

This work was carried out in collaboration between both authors. Both authors read and approved the final manuscript.

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ABSTRACT

Monitoring of ambient air quality parameters (NO₂, SO₂, NH₃, PM₁₀ and PM_{2.5}) levels in the surrounding area of a fertilizer plant located at Aonla, (U.P.) India, was carried out using the respirable dust sampler. The specific objectives were the determination of annual and seasonal variability of levels of ambient air pollutants and their relationship with the fertilizer plant emissions. The results have shown a marked seasonal trend and temporal variability of pollutants levels in the study area, where the relative contribution of individual seasons towards variation of worst cases were computed for all the ambient air quality monitoring years and were used to quantify the most potent season in terms of having worst ambient air quality. Through the complete monitoring period, summer season (March to June) with characteristic high production rate in the fertilizer plant (especially in month of march) were found to be having the worst-case scenarios for all the selected years (2013-2015) with a frequency of 32-33%, followed by the winter season (December to February) where in about 28% of cases the prescribed limits of national ambient air quality standards (NAAQS) were violated. Except summer and winter seasons, the remaining monsoon (July to September) and post-monsoon seasons (October to November) were also contributing, though less drastically.

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1. INTRODUCTION

The disturbing expansion of atmospheric pollution has led many countries of the world to set up several laws and regulations essential for maintaining the ambient air quality and the obligatory standard emission levels [1]. In this perspective, several monitoring networks were made in different countries to carry out real time qualitative and quantitative monitoring about the characteristics of ambient air, with importance given to atmospheric pollution and air quality monitoring and forecasting [2]. Monitoring and assessment of ambient air quality is a method to validate the effectiveness of the control measures implemented and for early detection of potentially harmful changes in atmospheric composition [3] and can be defined as a systematic, long term assessment and estimation of air pollutant level in any region or in atmosphere [4]. Monitoring is a very complex process, as it requires the collection of data that allows for a resolution of the dynamic nature of air quality in terms of its spatial and temporal variation [5] and is usually done to characterize and illustrate air quality in areas where there are large emission sources like industrial clusters and/or where there are sensitive environmental receptors [6].

An efficient air quality monitoring involves the consideration of many complex physical, chemical, socio-economic, environmental and meteorological factors, which helps as valuable tools to be engaged in developing and establishing of an effectual air quality monitoring strategy [5], with one main objective of shielding people and environment from extreme exposure to atmospheric pollutants; thus monitoring plays a crucial role in the development and assessment of these control strategies. Evaluating the success or failure of pollution control programs basically depends on the accessibility of precise data about ambient concentrations of air pollutants [7], and this data is used to determine whether an area is attaining the air quality standards or not and is also used to generate or validate air pollution dispersion models and effective control strategies [8]. Murena [9] conducted the data collection and analysis from various air monitoring stations in urban area of Napels (Italy) between 2001-2002 and has developed and implemented a daily air pollution index in order to highlight the effects of air pollution on health status of population.

Biswas et al. [3] conducted an analysis of ambient air quality conditions over Delhi from 2004-2009 and reported increased pollutant concentration and year to year variation in the design value of criteria pollutants. Nagendra et al. [10] analysed the air quality of Bangalore during 1999-2005, using statistical data provided by several air quality monitoring stations and results reveal that concentrations values of pollutants SO₂ and NO_x have increased and exceeds the standard limits in some intersections and arterial streets, due to traffic growth in recent years.

Air quality deterioration is interrelated to the potential of the atmosphere to scatter pollutants and to energy production and consumption patterns in the area under study. In most air quality applications the main concern is the dispersion in the atmospheric boundary layer, next to the earth's surface that is controlled by the surface heating and friction and the overlying stratification [11]. Air dispersion modelling has been acknowledged as a promising approach to predicting outdoor spatial and temporal variations and behaviour of pollutants, through numerical algorithms that take into account atmospheric dispersion, chemical and physical processes with an endeavour to estimated concentrations of pollutants [12]. The main issues under consideration in air pollution modelling are the complexity of the dispersion, which is controlled by terrain and meteorology effects along with the scale of the potential effects [13].

There are now comprehensive literatures on the use of models for prediction of ambient air quality. Chatterjee [14] carried out a study in Mangalpur industrial estate, Raniganj having many sponge iron industries, emit pollutants like SPM, RPM (PM₁₀), SO₂, NO_x and solid waste and making use of remote sensing and GIS technique the pollution zone was been recognized and health data was been collected accordingly. Bandyopadhyay [15] had done a dispersion modelling in assessing air quality of industrial projects under Indian regulatory regime. Rahman et al. [16] evaluated the ambient air pollution trend in Klang valley, Malaysia from 2007-2011 and used the GIS and principal component statistical analysis to predict the spatial trend of air pollutants. Banerjee et al. [17] performed a source contribution assessment of ambient NO₂ concentration at integrated industrial estate, Pantnagar through simulation of

Gaussian Finite Line Source Model (GFLSM) and Industrial Source Complex Model (ISCST-3) and model performances were evaluated. Le and Oamh [18] had done an integrated assessment of brick kiln emission impacts on air quality using the Industrial Source Complex Short-Term (ISCST3) dispersion model for the year 2006-2007 and found that SO₂ was the most critical pollutant, exceeding the hourly National Ambient Air Quality Standards.

Efficiency of an air quality management strategy depends also on the choice of monitoring sites, as irrational selection cannot successfully reflect the description of air pollution. Before, selection was done based on observed or empirical judgement or based on distance of polluters to neighbouring inhabited areas and population density [19]. Although, selection of monitoring sites simply in leeward or downward direction in regards to point source do not resolve the purpose of the assessing the spatial and temporal variations of airborne pollutants [20]. In an air pollution monitoring programme, selection of monitoring locations can be made based on utmost coverage and infringement detection for monitoring single or multiple pollutants; spatial coverage and data validity; or most favourable resolution based on a utility function [6]. However, for an industrial location, if the prime intention is the exposure of maximum average concentration, then monitoring location would be over-concentrated and situated around the high average concentration spots, along with the utmost population coverage should also be kept in mind as inhabitant safety is a concern [19]. Thus, air quality monitoring sites for an industry should be planned taking into consideration the exposure area of the potential zone, topography of the area and persistence and occurrence of wind speed and direction [21]. In India, CPCB, India has set-up 298 air quality monitoring stations across the country, but there is still need of a highly structured monitoring network within the country [22].

2. MATERIALS AND METHODS

2.1 Site Selection

The present research was conducted in Indian Farmers Fertiliser Cooperative Limited (IFFCO), a fertilizer manufacturing plant based in Aonla (U.P.). Geographically Aonla fertilizer complex is 28 km southwest of Bareilly on Bareilly Aonla Road in the state of Uttar Pradesh, located at in the northern region of India and at longitude 28°

13' 34.87" N and latitude 79° 14' 50.63" E at an elevation of 165 m above mean sea level.

2.2 Climatic Conditions

The climate of study area is warm, subtropical with dry hot summer and cold winter. The average annual temperature in the area is 25.1°C, with average annual rainfall is 1037 mm. When compared with winter, the summers have much more rainfall, starting in the third or fourth week of June and lasts up to September, in form of monsoon rains. The driest month is November, with 2 mm of rain. In July, the precipitation reaches its peak, with an average of 321 mm, there is a difference of 319 mm of precipitation between the driest and wettest months. June is the warmest month of the year with temperature averages of 33.3 °C. At 15.0 °C average, January is the coldest month of the year. The variation in annual temperature is around 18.3°C. The weather parameters for the study period (January 2013-december 2015) were persued from the regional meteorological station of India Meteorological Department (IMD) located at Lucknow in state of Uttar Pradesh.

2.3 Selection of Ambient Air Quality Monitoring (AAQM) Locations

The site selection was based on importance of emission sources, sensitivity of receptors, predominant local activities and wind directions in the area. However, for the air pollution assessment and estimation study, importance were principally given to ensure suitable determination of the spatial and temporal variations of pollutants, coupled with probable evaluation of their overall mean pollutant concentrations surrounding IFFCO-Aonla. Based on the topography and meteorological conditions of the pre-defined study region, primarily a five ambient air quality monitoring location was selected in the IFFCO-Aonla industrial unit, out of these five location, four were located inside the industry area and the fifth one was located in the residential area, considering its significance to assess emissions from both the vehicular pollution from adjacent State Highway-33 coupled with industrial sector. Selection of ambient air quality monitoring location was significant in regard to assess the impacts of recent industrialization at surrounding residential and sensitive areas. 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.

2.4 Methodology for Monitoring of Pollutants

For the monitoring of NO₂, SO₂, NH₃, PM₁₀ and PM_{2.5}, Respirable dust sampler (RDS APM 460BL, Envirotech, New Delhi, India) was used along with Thermo Electrically Cooled Gaseous Sampler (APM 411TE, Envirotech, Delhi) that was attached with RDS to monitor the gaseous pollutants.

For the determination of SO₂ and NO₂ gaseous pollutants, the monitoring was done at a constant flow rate of 1 l/min by bubbling ambient air through the liquid absorbing medium, however the proved modified Jacob and Hochheiser method [23] with absorbing solution of sodium hydroxide and sodium arsenite was used for the determination of NO₂. The concentration of nitrite ion (NO₂) produced during sampling was determined colorimetrically by reacting the NO₂ ion with phosphoric acid (H₃PO₄), sulphanilamide and N-(1-naphthyl)-ethylenediamine dihydrochloride (NEDA) and measuring the absorbance of the highly coloured azo-dye at 540 nm spectrophotometrically.

Improved West & Gaeke method with Potassium-tetrachloro-mercurate (K-TCM) as absorbing medium by BIS [24] was used to determine ambient SO₂ concentrations. Ambient SO₂ were collected by bubbling ambient air through a solution of potassium tetracholomercurate (TCM). A dicholomercuro sulphito mercurate complex which resists oxidation by the oxygen in the air was formed. This complex was stable to strong oxidants such as ozone and oxides of nitrogen and therefore, can be stored for some time prior to analysis. The complex was made to react with pararosaniline and methylsulphonic acid and the absorbance of the solution was measured by suitable spectrophotometer at 560 nm.

For the determination of NH₃, the Indophenol method by CPCB [25] with absorbing solution of sulphuric acid along with phenol and sodium hypochlorite was used. Ammonia in the atmosphere is collected by bubbling a measured volume of air through a dilute solution of sulphuric acid to form ammonium sulphate. The ammonium sulphate formed in the sample is analyzed colorimetrically by reaction with phenol and alkaline sodium hypochlorite to produce indophenol, a blue dye. The reaction is

accelerated by the addition of sodium nitroprusside as catalyst and measured the absorbance of the solution at 630 nm on a spectrophotometer.

The total Particulate Matter monitoring was performed using Respirable Dust Sampler (RDS, aerodynamic diameter <10µm), at an average flow rate of 1.2 m³/min as prescribed in BIS [26]. The particulate matter was measured by GF/A Glass microfiber filter paper (8"×10" size) (Whatman, England) while total non-respirable dust was measured by collecting the heavier particles deposited through cyclone (in black box) enclosed inside the RDS. Adequate precautionary measures were taken during total PM monitoring and concentrations were calculated gravimetrically. For the measurement of PM, filter paper was conditioned in desiccator for 24-h and weighed on a balance (Precisa, Germany) with the sensitivity of 0.001 g, both before and after air quality monitoring. The conditioned and weighed filter paper was placed in cloth-lined envelope and taken for monitoring to avoid any possibilities of contamination and moisture absorption. The total PM monitoring was performed at an average flow rate of 1.2 m³/min. In order to maintain the specific flow rate, the manometer reading was taken 3-4 times in a day so that the flow rate variations were kept within 1.1-1.3 m³/min. Average flow rate was finally considered for computing total amount of air sampled. Air quality monitoring was done once in a week continuously for 24-h. Adequate preventive measures were taken to avoid any sort of moisture absorption to filter paper and concentrations of total PM were calculated gravimetrically.

The statistical analysis conducted with the collected data was carried out with the help of standardized statistical techniques. The emission air samples collected from 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.

In order to formulate association and comparison between pre-identified monitoring locations in respect of existing meteorological conditions, concentrations of air pollutants (PM₁₀, PM_{2.5}, SO₂, NO₂ and NH₃) i.e., from January, 2013 to December, 2015 were considered for analysis. The weather parameters for the study period were persued from the regional meteorological

station of India Meteorological Department (IMD) located at Lucknow in state of Uttar Pradesh. Statistical relationship between the monitored air quality parameters and meteorological variables were determined through regression analysis using Microsoft Excel Data Analysis Tool programme. Further, monthly average concentrations of air pollutants were analyzed in respect of meteorological factors to compute correlation coefficient.

3. RESULTS AND DISCUSSION

3.1 Relative Comparisons between Individual Ambient Air Quality Monitoring Years

Ambient air quality of different air pollutants with respect to individual monitoring years are compared in Figs. 1 to 5, to identify the existing trend of variations of air pollutants monitored during study period. Such comparisons were found helpful to be acquainted with the particular air pollutant in terms of average concentration in a particular ambient air quality monitoring year and to get familiar with the existing climatic conditions prevalent during that particular year, with most deteriorated air quality.

a. SO₂

Ambient SO₂ concentrations for all pre-identified locations and years were found to have persisted within a narrow range of 4 - 7µg/m³. The annual average concentrations were also in control with respect to NAAQS. However, most particularly, SO₂ concentrations exhibited temporal pattern of declination in summer for monitoring years 2013 and 2014 and increasing pattern in most of the months in 2015. Though, the concentrations pattern for all monitoring years might not seem to be only influenced by the presence of fertilizer plant and/or local domestic sources but also the wind direction and speed coupled with atmospheric stability during monitoring period which might have been one reason for such observations [4].

b. NO₂

Ambient NO₂ concentrations measured in terms of NO_x for all selected locations and years were found to have persisted within a range of 4.2-7.6 µg/m³. The annual average concentrations were also found to be below the range with respect to NAAQS. On the other hand, most predominantly,

NO₂ concentrations depicted a temporal pattern of declination during the summer months for all the monitoring years 2013-2015 and showed an out of the ordinary pattern in rest of the months of years 2013-2015, with rising concentrations during monsoon in year 2014-2015 and a steep drop-n-rise in concentrations during year 2013. Entire monitoring periods were assumed to have influenced by changes in climatic conditions may be a pattern related to climate change and/or increased traffic emissions from nearby road and high production rate in factory and thus higher concentrations were expected [27].

c. NH₃

Ambient NH₃ concentrations for all pre-identified locations and years were found to have mostly persisted within a range of 15-25 µg/m³. The annual average concentrations were also in control with respect to NAAQS. However, most primarily, NH₃ concentrations exhibited temporal pattern of decrease during the summer months for all the monitoring years 2013-2015 and unusual pattern rise-n-drop during monsoon and winter months of years 2013-2015. One foremost reason for such observation may be prominent wind speed during the summer which leads to spreading of pollutants and the high temperature and scattered rainfall during the rainy season and high rate of production in fertilizer plant in winters which lead to NH₃ concentrations [28,29].

d. PM₁₀

Ambient PM₁₀ concentrations for all pre-selected locations and years were found to be persisting within a range of 150-200 µg/m³. The annual average concentrations were also not within the range prescribed by NAAQS which is a matter of concern in respect to health. Moreover, PM₁₀ concentrations shows temporal outline of decrease during the monsoon months for all the monitoring years 2013-2015, which may be due to adequate rainfall during monsoon and elevated levels of PM₁₀ concentrations were found in all AAQM years during winters which were anticipated due to persistence of atmospheric stability and reduced ventilation coefficient [4,30]. However, distinguishing factor was the presence of higher PM₁₀ concentrations during the pre-summer and summer months, may be due to generation of wind-blown particles from neighboring agricultural land.

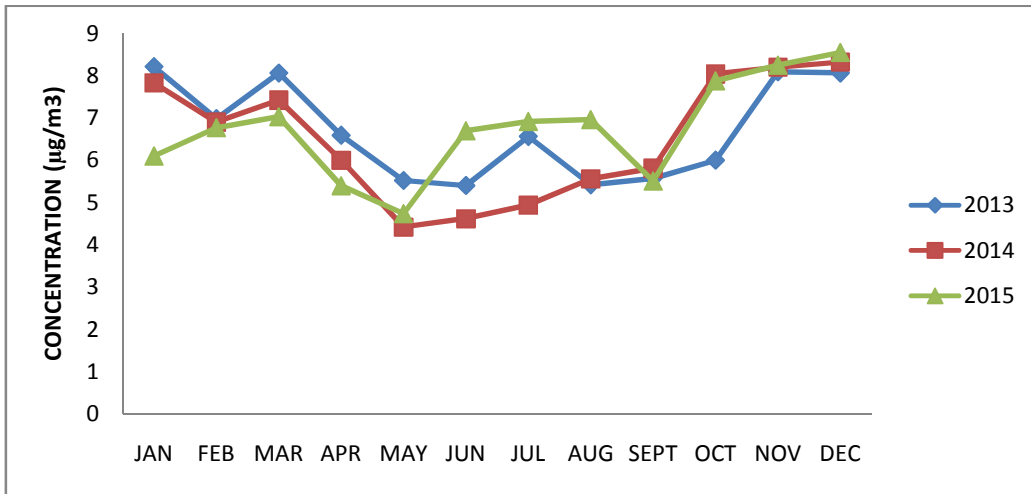


Fig. 1. Relative comparisons of SO₂ concentrations (µg/m³) between AAQM years

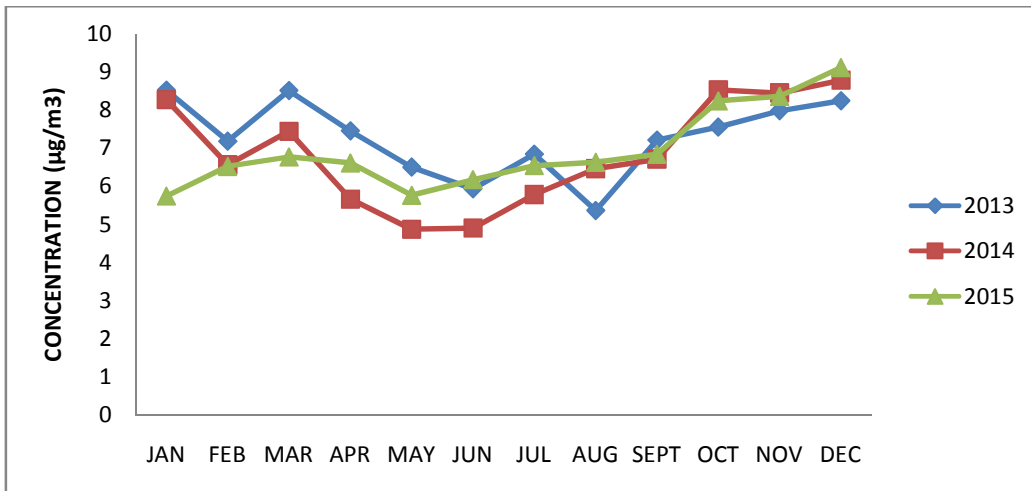


Fig. 2. Relative comparisons of NO₂ concentrations (µg/m³) between AAQM years

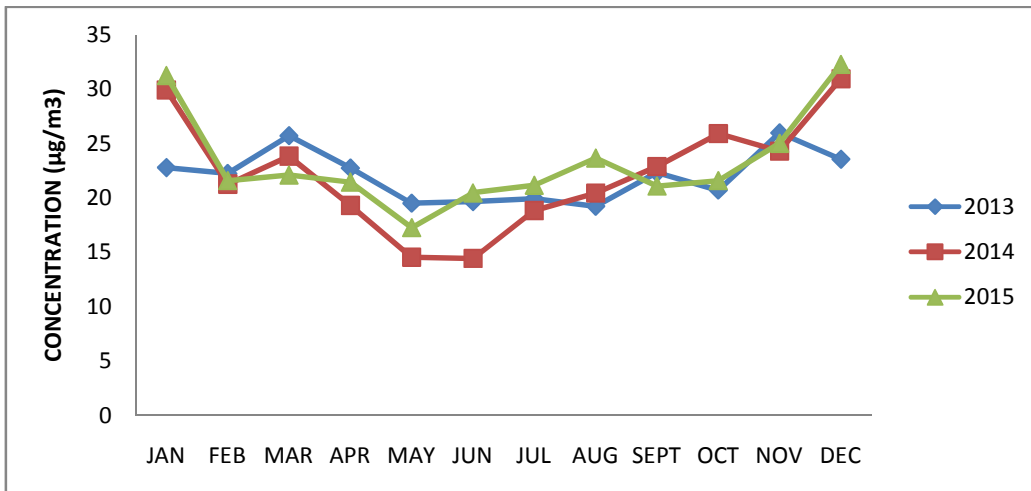


Fig. 3. Relative comparisons of NH₃ concentrations (µg/m³) between AAQM years

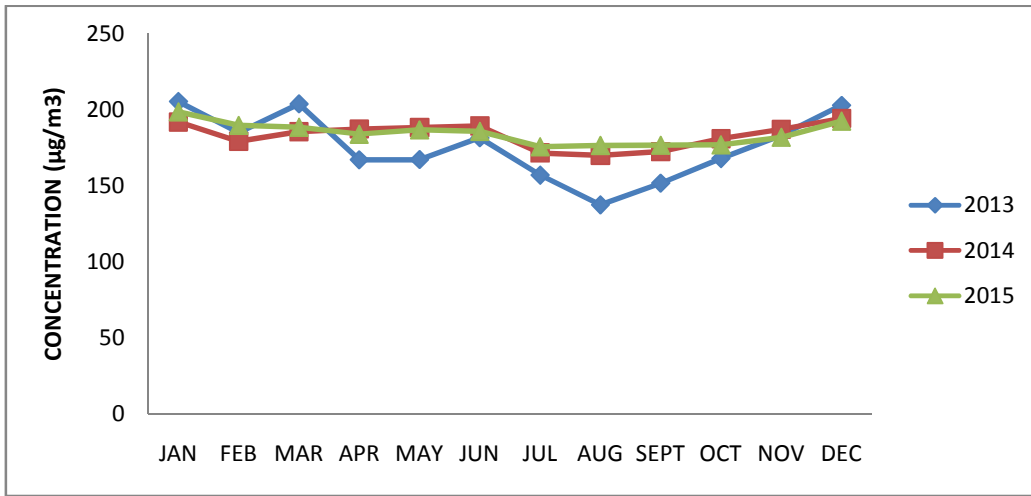


Fig. 4. Relative comparisons of PM₁₀ concentrations (µg/m³) between AAQM years

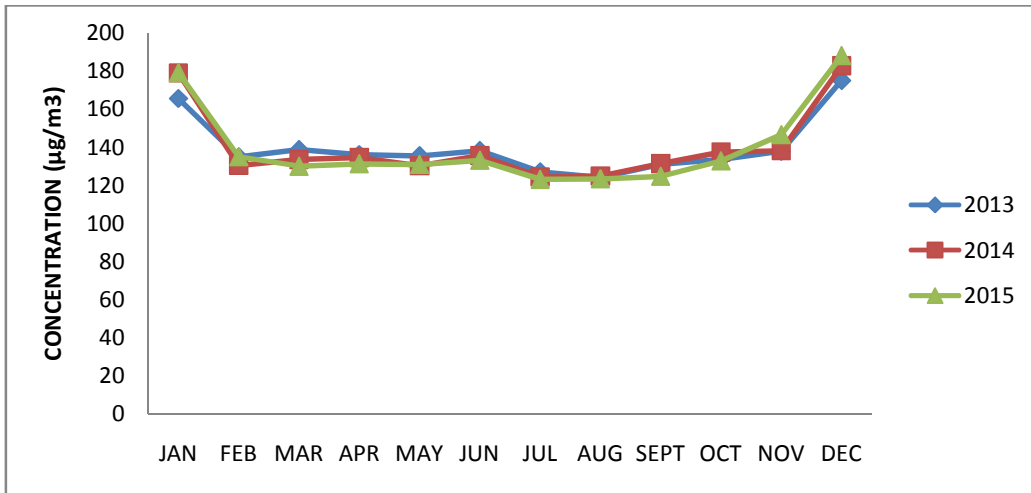


Fig. 5. Relative comparisons of PM_{2.5} concentrations (µg/m³) between AAQM years

e. PM_{2.5}

Ambient PM_{2.5} concentrations for all selected locations and years were found to exist mostly within the range of 120-140 µg/m³. The annual average concentrations were also higher than the range prescribed by NAAQS which can pose a substantial health effect. Moreover, PM_{2.5} concentrations shows temporal sketch out of declination during the monsoon period for all the monitoring years 2013-2015, which may be due to wind pattern and adequate rainfall during monsoon and the elevated levels of PM_{2.5} concentrations were found in all AAQM years during winters which were probable due to

prevalent atmospheric conditions as well as low wind speed leading to less distribution of particulate matter [6]. However, the presence of higher PM_{2.5} concentrations during the pre-summer and summer period was observed and it can be suggested that a large fraction of PM_{2.5} consists of soil-originated particles and because of re-suspension process, which is enhanced in dry climates, leads to such elevated concentrations. Considering resemblance between PM_{2.5} and PM₁₀, it may be concluded that PM_{2.5} concentrations for a particular year may perceived as a good indicator for prevalent concentrations of PM₁₀. Such findings were supported by [31,32,33,34].

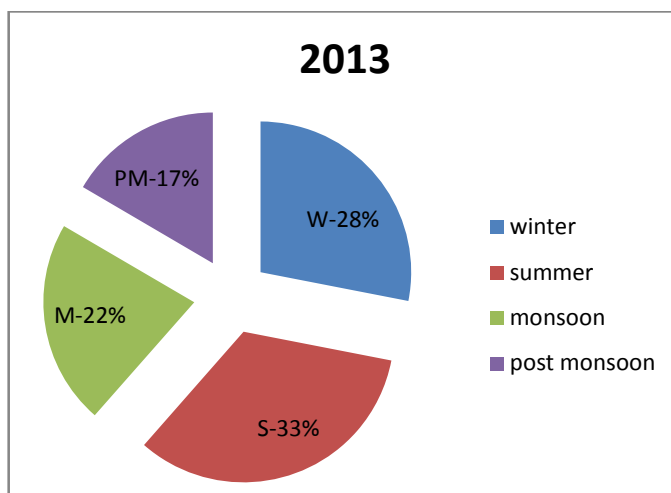
3.2 Relative Comparisons between Different Seasons

Relative contribution of individual seasons towards variation of worst cases (where pollutant concentration exceeds respective NAAQS) were further computed for all ambient air quality monitoring years and illustrated in Fig. 6. Such comparisons were used to quantify the most potent season in terms of having worst ambient air quality. A three year ambient air quality monitoring data (January 2013 to December 2015) were exclusively considered for significant appraisal of comparisons between different seasons. Through the complete monitoring period, summer (S) season (March to June) with characteristic high production rate in the fertilizer plant (especially in month of march) were found to be having the worst-case scenarios for all the selected ambient air quality monitoring years (2013-2015) with a frequency of 32-33%, followed by the winter (W) season (December to February) where in about 28% of cases the prescribed limits of NAAQS were violated. Except summer and winter seasons, the remaining monsoon (M) (July to September) and post-monsoon (PM) seasons (October to November) were also believed to contribute, though less drastically.

In year 2013 (January to December), it was observed that in nearly 33% of cases, concentrations were exceeding the NAAQS during summers. Moreover, winters contributes 28% of total worst air quality due to persistence of elevated PM concentrations. Apart from summer and winter seasons, the lingering monsoon (22%) and post-monsoon seasons (17%) were also found to contribute less

significantly. During the monitoring period of January, 2014 to December, 2014, it was found that winter and summer seasons both were contributing to the deterioration of air quality with a frequency of 28% and 33% respectively, followed by the remaining monsoon (22%) and post-monsoon (17%) seasons which were also contributing considerably in changing the ambient air quality status of the region. An identical trend was observed in year 2015 where summer contributes about 32% of cases when one or more pollutants exceeded the prescribed standards. The generations of worst cases in year 2015 were supposed to be higher during winter season (28%) with respect to monsoon (23%) and post-monsoon (17%) seasons contributing extensively.

The most substantial factors related to the observed results point mainly towards the dilution and dispersion of air pollutants, speed of wind, precipitation rate and the extent to which the emission can rise into the atmosphere (mixing height), as the probable reasons for concentrations obtained during the study period. The general meteorology of the study region during the winter is subjected to high pressure causing increased atmospheric stability, which in turn allows for less general circulation and thus more stagnant air [35]. Therefore, during winters, dispersion of atmospheric pollutants remains typically at its minimum and consequently elevated levels of pollutant concentrations are achieved [36]. On the contrary, during summers, the production rate of fertilizer in IFFCO plant was very high which lead to the more emission of pollutants and thus high concentrations observed, despite the average mixing height which remains at its highest naturally, resulting



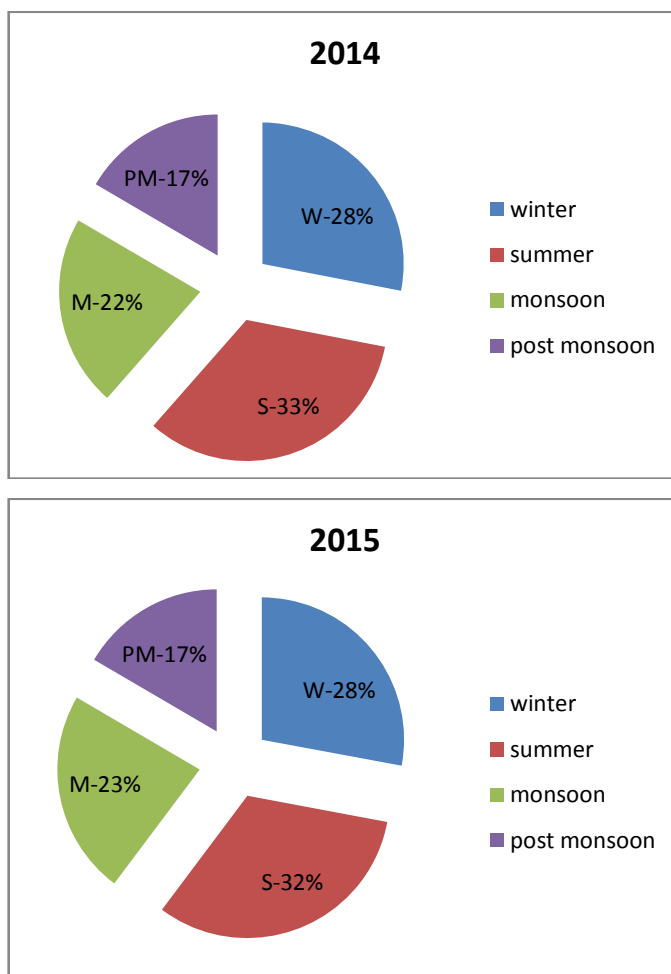


Fig. 6. Relative comparison of air pollutants within different seasons

mixing through a greater volume of air and hence resulting in lower pollutant concentrations. Furthermore, reduction of precipitation during monsoon and post-monsoon seasons reduces the potential for wet deposition and association cleaning mechanisms which might have lead to such elevated concentrations [37-40].

4. CONCLUSION

Relative contribution of individual seasons towards variation of worst cases (where pollutant concentration exceeds respective NAAQS) were computed for all ambient air quality monitoring years and were used to quantify the most potent season in terms of having worst ambient air quality. Through the complete monitoring period, summer season (March to June) with characteristic high production rate in the fertilizer plant (especially in month of march) were found to be having the worst-case scenarios for all the

selected ambient air quality monitoring years (2013-2015) with a frequency of 32-33%, followed by the winter season (December to February) where in about 28% of cases the prescribed limits of NAAQS were violated. Except summer and winter seasons, the remaining monsoon (July to September) and post-monsoon seasons (October to November) were also contributed, though less drastically. In year 2013, summer contributes about 33% of total worst air quality cases, followed by winters which contribute 28% of total cases due to persistence of elevated PM concentrations. In year 2014, winter and summer seasons both were contributing to the deterioration of air quality with a frequency of 28% and 33% respectively. An identical trend was observed in year 2015 where summer contributes about 32% of cases when one or more pollutants exceeded the prescribed standards. The generations of worst cases in year 2015 were supposed to be

higher during winter season (28%) with respect to monsoon (23%) and post-monsoon (17%) seasons contributing extensively.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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