

## Influence of Meteorology on Surface Ozone, NO<sub>2</sub> and Fine Particulate Matter at a Tropical Site in India

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#### Abstract

Regular measurements of surface Ozone, NO<sub>2</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> at Agra (27°10' N, 78°05' E, 169 msl), India have been carried out from Jan'11 to Dec'11 using online O<sub>3</sub> and NO<sub>x</sub> analyzers while PM<sub>2.5</sub> and PM<sub>10</sub> were collected using high volume sampler. Simultaneously, temperature, solar radiation, wind speed and relative humidity were also recorded using automatic weather station (WM 251). The diurnal variation of O<sub>3</sub> showed noontime maxima and NO<sub>2</sub> displayed its maxima in morning and evening hours. The seasonal variation of O<sub>3</sub> and particulate matter (PM) showed summer and winter maxima respectively. The relationship between atmospheric pollutants and meteorological parameters was studied using correlation analysis. O<sub>3</sub> showed significant positive correlation with temperature and solar radiation. NO<sub>2</sub> was found to be moderately correlated with wind speed while PM<sub>2.5</sub> and PM<sub>10</sub> were found to be significantly negatively correlated with temperature. Back trajectory analysis revealed that during summer and monsoon particulate matter at the site was contributed from distant sources while during the winter local sources appeared to dominate.

**Keywords:** Ozone, Oxides of Nitrogen, Particulate Matter, Meteorology, Back Trajectory

#### Introduction

The increasing concentrations of particulate matter (PM) and tropospheric ozone (O<sub>3</sub>) in the ambient air are responsible for decreasing visibility in a region. Studies have established that exposure to ozone and particulate matter is harmful to human health (NRC 1998). Particulate matter (PM) occurs in the atmosphere due to emissions from a number of anthropogenic sources like gasoline and diesel powered vehicles, combustible fuels such as coal, wood, gas and oil, various industrial processes and volatilization from polluted ground. The main concern for particulate matter is also due to the presence of polycyclic aromatic hydrocarbons (PAH) possessing carcinogenic and mutagenic properties. O<sub>3</sub> is regarded as a powerful green house gas in the atmosphere after water vapor, CO<sub>2</sub>, CH<sub>4</sub> and nitrous oxide with a radiative forcing of +0.35 Wm<sup>-2</sup> (IPCC 2001). O<sub>3</sub> is both cyto-toxic and phyto-toxic in nature. One molecule of ozone produced in the atmosphere is 1200-2000 times more effective in global warming than an additional CO<sub>2</sub> molecule (Lal 2007). It is a secondary pollutant photo chemically produced during the oxidation of methane (CH<sub>4</sub>), carbon monoxide (CO) and non-methane volatile organic compounds (NMVOC) in the presence of nitrogen oxides (NO<sub>x</sub> = NO+NO<sub>2</sub>) (Tang et al. 2009). The five main factors, which control the tropospheric ozone levels, are photo-chemical production, chemical destruction, long-range transport, surface dry deposition on vegetation, water and other materials and stratospheric-tropospheric exchanges (Alvim et al. 2010). Due to strong association between air pollutant levels and meteorological conditions, meteorological variability influences the air quality caused by changes in emissions of pollutants (Rao and Zurbenko 1994). Additionally chemical generation of pollutants and their deposition on the surface are strongly affected by meteorological variables (Tarasova et al. 2003). With this view, the influence of local meteorological conditions on tropospheric pollutants (tropospheric ozone, NO<sub>2</sub> and particulate matter, PM<sub>2.5</sub> and PM<sub>10</sub>) levels was studied at Dayalbagh, Agra from January 2011 to December 2011. In this study, an attempt has been made to address briefly the diurnal-seasonal variations in surface ozone, NO<sub>2</sub> and particulate

matter and the inter-relation of these pollutants and available meteorological parameters (temperature, wind speed, relative humidity).

## Materials and Methods

### Sampling Site

Agra (27°10' N and 78°05' E) with an altitude of 169 m above sea level lies in the semi-arid zone of India and is positioned with the Thar Desert of Rajasthan to the West, central hot plains to the South, Gangetic plains to the East and cooler hilly regions to the North (Figure 1).



**Figure 1: Map showing geographical location of the sampling site**

Sampling was performed at Dayalbagh, a sub-urban area located in the north of Agra primarily a vegetated area with minimum industrial activity. The sampling site lies by the side of a road that carries mixed vehicular traffic, moderate (of the order of 1000 vehicles) during the day and minimal (of the order of 100 vehicles) at night. The campus lies about 2 km north of the National Highway-2 (NH-2), which has dense vehicular traffic ( $10^6$  vehicles) throughout the day and night (Singla et al. 2011).

### Measurement of Ozone and NO<sub>x</sub>

Measurement of surface Ozone and NO<sub>2</sub> was carried out at Dayalbagh from January 2011 to December 2011. O<sub>3</sub> concentrations were recorded using continuously operating O<sub>3</sub> analyzer (Thermo Fischer Model 49i) based on ultraviolet absorption photometry, resting upon absorption of radiation with the wavelength of 254 nm by ozone in the analyzed sample. Calibration of the system was done regularly with the help of a built-in ozone generator. Zero setting of the analyzer was also performed regularly by admitting zero air (free of ozone) into the analyzer. The minimum detection limit and precision of the analyzer are about 0.5 ppbv and 1 ppbv (2 %) respectively. NO<sub>x</sub> concentrations were recorded with a NO<sub>x</sub> analyzer (Thermo Fischer model 49i) based on the chemiluminescence effect of NO<sub>2</sub> produced by the oxidation of NO by O<sub>3</sub> molecules, which peaks at 630 nm radiation. In this analyzer, NO<sub>2</sub> is measured by converting it into NO using the thermal conversion (heated molybdenum) method. The minimum detection limit and precision of NO<sub>x</sub> analyzer are 0.4 ppbv and 1 ppbv (2%) respectively. Calibration for NO<sub>2</sub> was done by using permeation tube (KINTEK, TEXAS). The online O<sub>3</sub> and NO<sub>x</sub> analyzers record

concentrations on an averaging time of every 5 minutes. The 5 minute average concentrations were further averaged to get one hour average O<sub>3</sub> and NO<sub>x</sub> concentration (Singla et al., 2011)

### Sampling of PM<sub>2.5</sub> and PM<sub>10</sub>

Concurrently PM<sub>2.5</sub> and PM<sub>10</sub> were collected using a high volume sampler, Envirotech APM 550, installed on the roof of a building. The samples were collected on pre-desiccated and pre-weighed glass fiber filters (Whatman, EPM 2000). Air was drawn at a flow rate of 16.6 L min<sup>-1</sup> for 24 hr on an average for collection of both PM<sub>2.5</sub> and PM<sub>10</sub>. Each filter was weighed at least thrice before and after sampling and the net mass was obtained by subtracting the pre-sampling weights from the post-sampling weights. PM concentration was calculated by dividing the net mass by the volume of air sampled.

### Recording of Meteorological Parameters

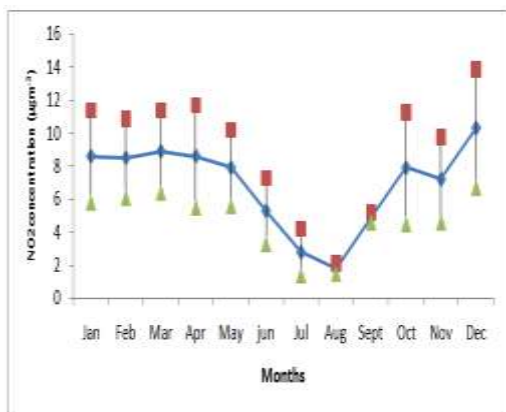
Ambient temperature, solar radiation, relative humidity, wind speed, and wind direction were recorded through an automatic weather monitoring system (WM251) mounted on the roof. It was programmed to collect data at 1 min interval and store them in memory to be downloaded to a computer. In summer months, the average temperature ranged from 40-45°C and average humidity varied from 25-50%. Summer was also associated with strong dust storms (wind speed varied from 10-16 kmh<sup>-1</sup>). The average temperature varied from 22-28°C and relative humidity varied from 70-75% in winter months. During monsoon (July-September) relative humidity reached up to 80-85%.

## Results and Discussion

### Trends of Ozone and NO<sub>2</sub> Concentrations

Figure 2 displays the mean and 1σ standard deviation of O<sub>3</sub> and NO<sub>2</sub> concentrations observed in each month during the study period (30/31 day average). The highest monthly mean O<sub>3</sub> concentrations were observed in summer season (April to June) followed by winter (December to February), post monsoon (October and November) and monsoon (July to September). The higher ozone concentrations varied from 96.5 to 129.3 μg m<sup>-3</sup> during summer season while the lower O<sub>3</sub> concentration varied from 11.9 to 31.9 μg m<sup>-3</sup>.





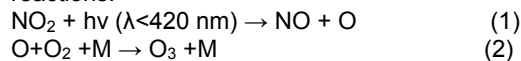
**Figure 2: Mean Concentration levels of O<sub>3</sub> and NO<sub>2</sub> in different months**

On the other hand during winter and post monsoon season, the average O<sub>3</sub> concentration (stretched from 46.04 to 96.79 µg m<sup>-3</sup> in winter and 55.2 to 91.4 µg m<sup>-3</sup> in post monsoon) was observed to be much lower than that observed in summer season. This might be due to peak surface temperature observed in summer reaching as high as 41 to 46°C and lowest seasonal total rainfall of 30.9 mm. The maximum O<sub>3</sub> concentration varied from 154.2 to 210.2 µg m<sup>-3</sup> and minimum concentration varied from 8.78 to 21.6 µg m<sup>-3</sup> in winter season. During post monsoon, the maximum concentration varied from 139.8 to 208.1 µg m<sup>-3</sup> and minimum varied from 7.71 to 20.7 µg m<sup>-3</sup>. Low concentrations of O<sub>3</sub> were observed in monsoon season (average concentration varied from 43.5 to 70.2 µg m<sup>-3</sup>). During monsoon season, the maximum levels varied from 108.7 to 146.3 µg m<sup>-3</sup> and minimum levels varied from 4.7 to 12.2 µg m<sup>-3</sup>. These lower concentrations are attributed to scavenging of pollutants by rain. (Reddy et al., 2008). Similarly, higher concentrations of NO<sub>2</sub> were observed in summer, winter and post monsoon season with remarkably lower values in monsoon season. The average NO<sub>2</sub> concentration varied between 12.4 to 17.8 µg m<sup>-3</sup> in summer, 10.3 to 14.1 µg m<sup>-3</sup> in winter, 7.6 to 11.5 µg m<sup>-3</sup> during post monsoon and 4.4 to 6.5 µg m<sup>-3</sup> in monsoon season. Relatively higher concentrations observed in winter might be due to temperature inversion phenomenon while lower concentrations in monsoon could be associated with rains and overcast sky (50–100%) conditions (www.wunderground.com).

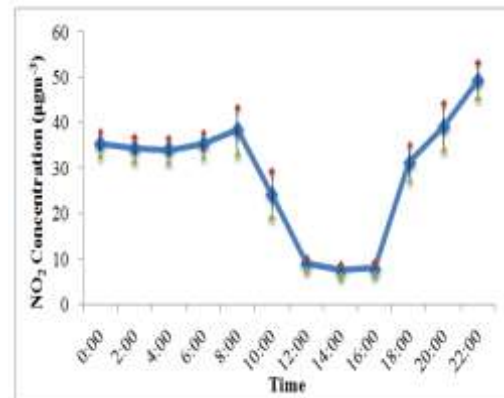
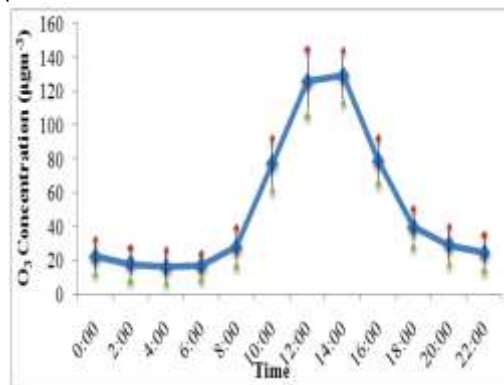
**Diurnal Variation**

The variation of surface ozone, within a day, may be helpful in delineating the processes responsible for ozone formation at a particular location. A representative average diurnal pattern of O<sub>3</sub> and NO<sub>2</sub> is shown in Figure 3. As expected, time concentration profiles of O<sub>3</sub> show a potential and direct link with NO<sub>2</sub> variability. In general, O<sub>3</sub> concentration starts increasing gradually after sunrise (0800 h), attains maximum concentration at noon hours and then gradually decreases and thus closely follows the diurnal cycle of solar radiation and surface temperature. However, NO<sub>2</sub> concentration shows a sharp increase and reaches a maximum at a time that

coincides with the maximum automobile traffic in the morning and evening respectively and thus follows an inverse relationship with O<sub>3</sub>. In the early morning hours before sunrise (0600 h), O<sub>3</sub> levels are observed to be very low and almost constant (16–22 µg m<sup>-3</sup>). With the onset of sunshine, as solar radiation increases, O<sub>3</sub> concentration gradually increases (39–79 µg m<sup>-3</sup>) probably due to its photochemical formation through photolysis of NO<sub>2</sub> via following set of reactions.



The concentration of O<sub>3</sub> that attains maximum values (125–129 µg m<sup>-3</sup>) at noontime (1200–1400 h) can be attributed to O<sub>3</sub> production by photooxidation of precursor gases, like CO, CH<sub>4</sub>, and other hydrocarbons in the presence of sufficient amount of NO/NO<sub>x</sub>. In the evening hours, a decrease in the O<sub>3</sub> values (24–29 µg m<sup>-3</sup>) is observed probably due to lower production of O<sub>3</sub> as well as its depletion due to the fast reaction with NO forming NO<sub>2</sub> (O<sub>3</sub> + NO → NO<sub>2</sub> + O<sub>2</sub>). This daytime increase in ozone concentration is a pronounced feature of an urban site (Lal et al. 2000, Reddy et al. 2008). Boundary layer processes and meteorological parameters also play a role in the O<sub>3</sub> variability. After sunrise, boundary layer height gradually increases from about 200–300 m to about 1500–2000 m during noon hours due to convective heating, and the layer becomes stratified (Reddy et al. 2008b). The photochemical production of O<sub>3</sub> is directly related to temperature and solar flux, its concentration increases with respect to both parameters.



**Figure 3: Average Diurnal Variation of O<sub>3</sub> and NO<sub>2</sub>**

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NO<sub>2</sub> mixing ratios are generally stronger during the morning and late evening hours. Variations in NO<sub>2</sub> are caused by variations in boundary layer mixing processes, chemistry, anthropogenic emissions, and local surface wind patterns (Rao et al. 2002a Rao et al. 2002b). The emitted pollutants get trapped at the lower heights in the boundary layer during evening and continue to remain so until early morning due to formation of a nocturnal stable layer. However, pollutants are diluted during midday due to increased height of the boundary layer and extensive mixing. Vehicular emissions serve as the dominant source for NO<sub>2</sub> in this region. During the morning hours (0600–0800 h) the high values of NO<sub>2</sub> (39–45  $\mu\text{g m}^{-3}$ ) have been observed and hence expected O<sub>3</sub> titration to become faster. This might be because of increase in heavy-duty diesel truck activities that serves as a significant source of NO<sub>x</sub> emissions. At evening traffic peak hours (2000 h), a secondary NO<sub>2</sub> maximum (~39.3  $\mu\text{g m}^{-3}$ ) is observed during which time O<sub>3</sub> exhibits a secondary minimum (~7.5–9  $\mu\text{g m}^{-3}$ ). A diurnal pattern with maximum ozone concentration during daytime and minimum during nighttime is also observed during dry months at various locations in India like Ahmedabad, Delhi, Pune, Trivandrum, Anantapur and Gadanki (Purkait et al. 2009, Naja et al. 2003, Satsangi et al. 2004, Sahu and Lal 2006, Londhe et al. 2008, Debaje and Kakade 2009, Naja and Lal 2002).

## PM<sub>2.5</sub> and PM<sub>10</sub>

The mass concentrations of both PM<sub>2.5</sub> and PM<sub>10</sub> exhibited distinct seasonal variations similar to many other areas in the world (Zheng and Fang 2000). The mass concentrations displayed higher concentrations in winter than monsoon by a factor of 2. Figure 4 shows the monthly variation in PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations. As observed from the graph, the mass concentration of PM<sub>2.5</sub> varies between 58–84  $\mu\text{g m}^{-3}$  in summer season (followed by a significant dip in concentrations in monsoon season where mass concentration varied from 23–32  $\mu\text{g m}^{-3}$ ). Thereafter, the mass concentrations start rising rapidly in October and become highest in winter season with the concentration reaching between 105–217  $\mu\text{g m}^{-3}$ . Likewise the mass concentration of PM<sub>10</sub> varied from 178–250  $\mu\text{g m}^{-3}$  in summer season followed by lower concentrations (42–79  $\mu\text{g m}^{-3}$ ) in monsoon and markedly higher concentrations (110–324  $\mu\text{g m}^{-3}$ ) in winter.

The seasonal distribution of particulate matter is proportionately governed by the combined effect of emission factors, dispersion conditions and chemical mechanisms. In Agra (India), summers are characterized by dry climate with intense dry storms and intermittent rainfall is observed in the monsoon while winters are usually associated with calm periods and stagnant atmospheric conditions. During winters, particulate emissions from automobile exhaust are higher due to low ambient temperature and traffic congestion due to poor visibility, fog, mist and haze. Also fuel combustion processes involving wood burning and coal combustion for cooking and space heating during winter add to the levels of particulate

matter. The lower particulate concentrations in monsoon are attributed to their scavenging through precipitation and reduced soil re-suspension. The sources and transport pathways of particulate matter were identified using air mass backward trajectory analysis. These air mass back-trajectories were calculated using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model (<http://ready.arl.noaa.gov/HYSPLIT.php>) which is based on the GDAS Meteorological Data (Draxler et al. 2003). The five-day back trajectories for different seasons (summer, monsoon and winter) were calculated at 14:00 h (local time) for 1000, 1500 and 2000 m above the ground level and have been represented in Figure. 6.

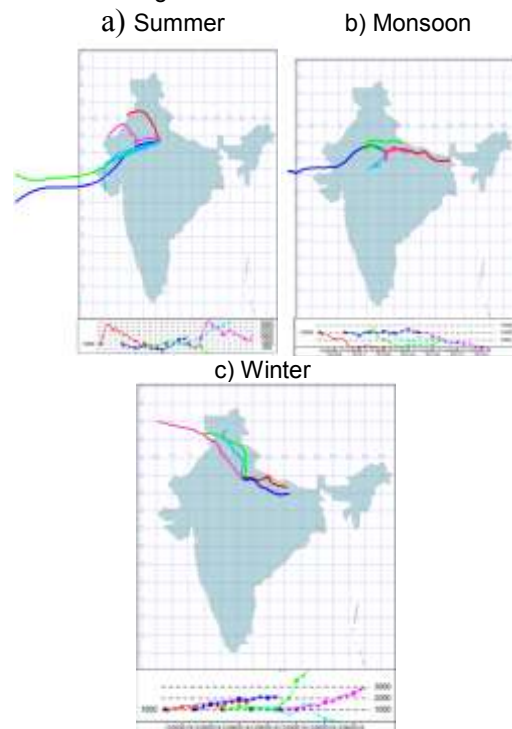
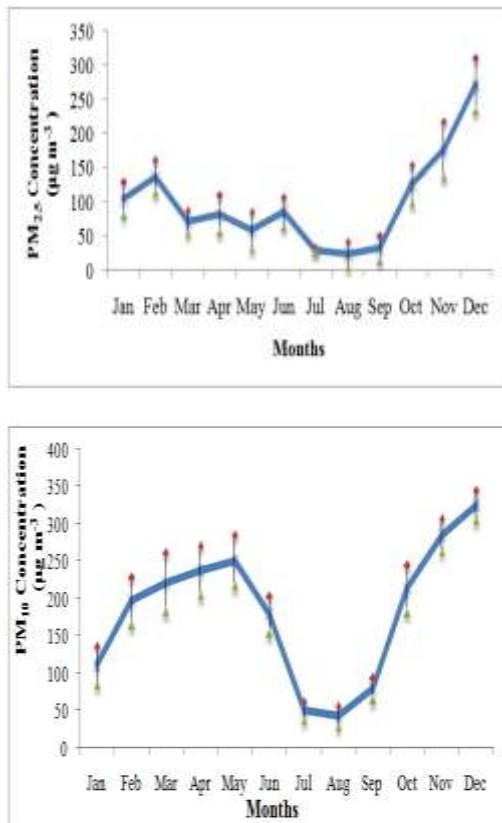


Figure 6: The typical air mass five-day trajectories arriving at the sampling site in a) summer season pointing to the dust source from northwest Asian desert regions b) monsoon season indicating the origin of air masses from the Arabian Sea and Bay of Bengal and c) winter season indicating local origin of aerosols

In summer long-range transport of dust from dust storms originating from the Thar Desert contributes to higher concentrations of particulate matter. During monsoon season, the moisture loaded winds originating from Arabian Sea and Bay of Bengal causes heavy rainfall, thereby lowering particulate concentrations significantly. Short trajectories originating from shorter distances in winter season reflects the major contribution from local sources to higher concentrations of PM<sub>2.5</sub> and PM<sub>10</sub>.

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**Figure 4: Monthly mean concentration of PM2.5 and PM10**

### Relationship of Tropospheric Pollutants with Meteorological Parameters

#### Relationship of O<sub>3</sub>, NO<sub>2</sub> and Particulate Matter with Temperature

The temperature variance observed during the study period was found enough to study the relationship between temperature and pollutants of interest. This was studied on a seasonal basis as shown in Table 1. The correlation between temperature and primary pollutant, NO<sub>2</sub> was found to be insignificant in all seasons as compared to secondary pollutant, O<sub>3</sub>. Surface ozone, an indicator of photo-oxidation processes exhibited positive trend with surface temperature. O<sub>3</sub> increases significantly with the increase in surface temperature and showed strong positive correlation with temperature in summer and a moderate correlation in winter (Table 1). Both PM<sub>2.5</sub> and PM<sub>10</sub> showed an inverse correlation with temperature (Table 1). This negative correlation could be attributed to dispersion of particulate matter due to unstable conditions associated with high temperature as well as due to faster thermal degradation of some constituents of aerosols. In general, higher concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> were observed during days with temperature less than 20°C.

**Table 1: Correlation between Pollutants and Meteorological Variables**

	Season	Temp	WS	RH
O <sub>3</sub>	Summer	0.75*	0.52*	-0.65*
	Winter	0.57*	0.48*	-0.82*
	Monsoon	0.45	0.29	-0.26
NO <sub>2</sub>	Summer	-0.43	0.55*	-0.49*
	Winter	-0.55	0.67*	-0.31
	Monsoon	-0.20	0.26	-0.25
PM <sub>2.5</sub>	Summer	-0.59*	-	0.74*
	Winter	-0.56*	-	0.81*
	Monsoon	0.12	-0.36	0.50*
PM <sub>10</sub>	Summer	-0.62*	-	0.68*
	Winter	-0.58*	-	0.76*
	Monsoon	0.12	0.10	0.52*

Correlations are significant at 0.05 level (2 tailed)

#### Relationship of O<sub>3</sub>, NO<sub>2</sub> and Particulate Matter with Wind Speed

The prevailing winds loaded with moisture or aerosol particles from a distant source play a major role and may affect the tropospheric pollutant concentration. The concentration level of O<sub>3</sub> and NO<sub>2</sub> were observed to increase with the transport of wind and hence showed positive correlation with wind speed. O<sub>3</sub> was found to be moderately correlated with wind speed and NO<sub>2</sub> showed considerable positive correlation (Table 1). On the other hand, the negative relationship between particulate matter and wind speed suggest that strong winds flush away the particulates from the study area and low winds allow particulate levels to rise. Concentrations of PM<sub>2.5</sub> greater than 100 µg m<sup>-3</sup> were observed with wind speed less than 2 km h<sup>-1</sup>. Similarly, PM<sub>10</sub> concentrations were higher than 250 µg m<sup>-3</sup> when wind speed was less than 2 km h<sup>-1</sup>.

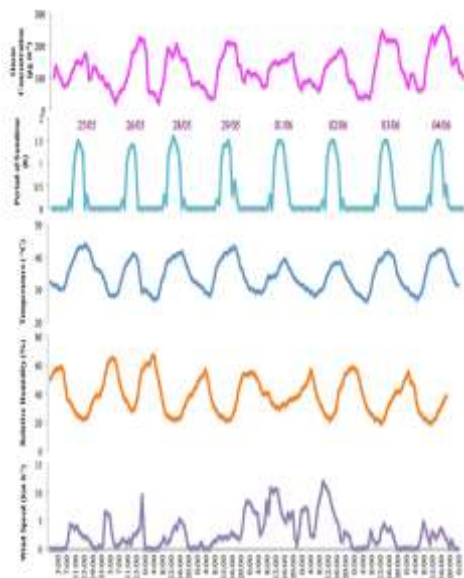
#### Relationship of O<sub>3</sub>, NO<sub>2</sub> and Particulate Matter with Relative Humidity

O<sub>3</sub> and NO<sub>2</sub> showed significant negative correlation with relative humidity (Table 1). This may be attributed to enhanced oxidation of hydrocarbons in the afternoon and thereby supports the ozone production (Ma and Van 2000). The reaction of NO<sub>2</sub> with OH is decreased at lower relative humidity and hence concentrations of NO<sub>2</sub> increase. On the other hand, PM<sub>2.5</sub> and PM<sub>10</sub> showed moderately positive correlation with humidity.

#### O<sub>3</sub> Episode

Agra is a tropical site, experiences extreme climatic conditions in summer season and in the winter season as well. The site observes continuous 7-9 hr of sunshine during a day i.e. from 8:00 a.m. in the morning till 5:00 p.m. in the evening. Since solar radiation is the major factor responsible for the

photochemical processes to occur, hence significantly high  $O_3$  concentrations were observed during nine consecutive days from 25<sup>th</sup> May to 4<sup>th</sup> June 2011, reaching as high as  $258 \mu\text{g m}^{-3}$ . Figure 5 plots the hourly variations of  $O_3$  concentration, period of sunshine, temperature, relative humidity and wind speed for 9 consecutive days identified as episodic days in the summer of 2011.



**Figure 5: Hourly variations in  $O_3$  concentration, period of sunshine, temperature, relative humidity and wind speed during high  $O_3$  days**

Being in the tropical zone, the maximum temperature ranged from 40–45 °C. As observed from the figure 5, low temperatures (28–32 °C) are observed in morning hours (6:00 – 8:00 a.m.) and thereafter starts rising and becomes maximum (45–46 °C) in noon hours (12:00 – 2:00 p.m.). Particularly, ground level ozone concentrations show positive correlation with surface temperature. Relative humidity varied from 25–60% during the episodic  $O_3$  days and shows significant negative relationship with  $O_3$  concentration.  $O_3$  destruction occurs whenever an excited oxygen atom  $O(^1D)$  reacts with  $H_2O$ . Additionally, water vapour also enhances the removal of short-lived radicals such as HOx, which are important precursors for  $O_3$  formation. Further, wind speed varied from 2.5 to 5.6  $\text{km h}^{-1}$  during night hours and 6.1 to 11.3  $\text{km h}^{-1}$  in the daytime hours i.e. from 10:00 a.m. to 4:00 p.m. Since low wind speed favors  $O_3$  formation, therefore wind speed was observed to be moderately correlated with  $O_3$  concentration.

#### Conclusion

The diurnal and seasonal variation of  $O_3$  displays noon-time ( $125\text{--}129 \mu\text{g m}^{-3}$ ) and summer season maxima ( $96.5$  to  $129.3 \mu\text{g m}^{-3}$ ) respectively while diurnal variation of  $NO_2$  shows morning and evening maxima ( $39\text{--}45 \mu\text{g m}^{-3}$ ) corresponding to traffic rush hours. The mass concentration of  $PM_{2.5}$  ( $105\text{--}217 \mu\text{g m}^{-3}$ ) and  $PM_{10}$  ( $110\text{--}324 \mu\text{g m}^{-3}$ ) was observed to be maximum in winter. Ozone was positively correlated with temperature while  $PM_{2.5}$  and  $PM_{10}$  were negatively correlated with temperature. Solar radiation was also strongly related with  $O_3$ .

Wind speed showed positive correlation with  $NO_2$ .  $O_3$  and  $NO_2$  showed significant negative trend with relative humidity respectively which may be due to enhanced oxidation of hydrocarbons in the afternoon.

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