

Long Term Measurement of Surface Ozone and Its Precursor's at Hyderabad - a Tropical Urban Site

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Abstract:The temporal variations of surface ozone (O₃) and its precursor trace gases were studied at tropical urban site of Hyderabad. Ozone and oxides of nitrogen (NO_x = NO+NO₂), and meteorological parameters were continuously monitored at the established air monitoring station. From the results it is observed that there substantial change in the concentrations of surface O₃ during different seasons of the year. Maximum mean concentrations of O₃ observed during summer. Diurnal-seasonal changes in surface O₃ and NO_x concentrations dependent on atmospheric chemistry, boundary layer dynamics, and local meteorological conditions. In addition, nocturnal chemistry of NO_x played an important role on the formation of O₃ during day time.

Key words: Ozone, Diurnal variation, Trace gas, Air pollutants, Photochemistry.

1. INTRODUCTION:

Rapid industrial development during past few decades in the Indian region has resulted in a tremendous growth in emission of pollutants in the atmosphere. Hence, studies related to the emissions of these pollutants, especially at surface level over tropical region, carried out in several sites in the Indian region. O₃ variations studies conducted at Ahmadabad, an urban site showed O₃ concentration upto 80 ppbv [1]. The ozone concentration invariably peaked at noontime and remained high during early summer and spring periods. It revealed that daytime O₃ production mainly is due to the photo-oxidation of its precursor gases. Additionally boundary layer process, surface wind patterns, meteorology also play the role on variations of pollutants at this site.

Study of surface ozone and its precursors at Dayalbagh, Agra located at Indo-Gangetic plain site during 2008-09 by Singla et al., found that the daytime maxima and pronounced maxima in summer and winter seasons and minima in monsoon and post monsoon seasons [2]. Varsheny and Aggarwal reported the amount of O₃ in the ambient air varied between 9.4 - 128.3 ppbv exhibiting wide temporal variation in Delhi [3]. Monthly 24 h mean O₃ concentration varied in the range of 6.12-34.68 ppbv at relatively clean area in Varanasi, India [4]. A study conducted by Agrawal et al., reported that O₃ concentrations (6hr mean) ranging from 10.3 to 15.4 ppbv during winters and from 9.7 to 58.50 ppbv during summers at different sites in Varanasi city [5].

Despite after noon maximum concentration of surface O₃ at urban or rural sites, measurement at Nainital, a high altitude site in the central Himalayas do not show the daytime photochemical build up [6]. Moreover, surface O₃ measurements at another high altitude site Mt Abu during the period 1993–2000 by Naja et al., demonstrate that O₃ mixing ratios showed a low value during day time and the diurnal pattern changes significantly after the northeast monsoon period [7]. Additionally, they found that local pollutants are major contributors for higher O₃ levels in late autumn and winter period, while regional transport plays a role in spring season. The monthly pattern of O₃ was found to be increased from January to May and thereafter started decreasing up to July at another Himalayan site located at Mohal, in Kullu Valley of the North western part of Indian during the year 2010 [8]. A maximum monthly mean level of O₃ observed was 84 ppbv at 16:00 hrs in May at this site.

Surface O₃ concentration in Delhi is found to be high enough to exceed “Critical levels” which are still considered safe for human health, vegetation and forest [9]. In addition, they found that accumulated exposure over a threshold of AOT40 of O₃ exceeded significantly during winter and pre-monsoon season in India. Studies on surface O₃ at a rural site (Ananthapur) by Reddy et al., showed distinct diurnal and seasonal variability and revealed that the rate of increase of O₃ is high in March and low in July [10]. Surface O₃ and precursor gases measurement at other rural site Gadanki by Naja and Lal during 1993–1996 observed a poor correlation between O₃ and NO_x

which indicates that NO_x levels at Gadanki are not controlled by fresh combustion or emissions and could be transported from the nearby major cities [11].

2. SITE DESCRIPTION

The experimental site was located at Hyderabad (17.170 - 17.830 N to 78.170 - 78.830E), (Figure 1). The trace gas measurements were carried out at

Hyderabad during the period 2010 to 2014 along with the meteorological variables. Hyderabad is situated in the Deccan plateau and has an average elevation of about 536 m above mean sea level. Hyderabad is one of the largest cities in India.



Fig 1: Geographical location of air monitoring station (blue).

The rate of urbanization in Hyderabad city is very rapid since 1960 making it the sixth most populous city. The population of Hyderabad urban population has drastically increased from 4.48 million in 1901 to 7.75 million in 2011 (<http://www.census2011.co.in/census/city/392-hyderabad.html>). The decadal growth of population has been decreased from (1991-2001) to (2001-2011). The reports by Census India 2011 (<http://www.census2011.co.in/census/district/122-hyderabad.html>) recorded the population density of 18,172 people per square kilometer for Hyderabad. As the city has grown up rapidly over the years the quality of environment has declined steadily. Rapid urbanization and increased economic activity has led to increased industrial waste, air, noise and water pollution.

In Hyderabad, the air pollution load is mainly contributed by anthropogenic emissions. The city comprises of many industrially developed areas and the site is surrounded by many industries such as Electronics Corporation of India Limited (ECIL), Hindustan Cables Limited (HCL), Nuclear Fuel Complex (NFC), petroleum storage containers, bottling units of Hindustan Petroleum Corporation Limited (HPCL), Bharat Petroleum Corporation Limited (BPCL) in the south and southeast directions. The emission sources at these sites are majorly contributed by automobile traffic (both light and heavy motors) and liquefied petroleum gas (LPG) leakage from the nearby gas filling stations. Guttikunda and Kopakka reported the sector-specific emissions (2010–2011) for this city, which accounted for 113,400 t of VOCs [12].

The air samples were simultaneously analyzed for surface O₃, NO_x, CO, and SO₂. Auto-sampling was made at 10 meters elevation above the ground level. The trace gases in ambient air were measured for every 5 min interval with Thermo Scientific instruments and the data were retrieved using Envidas® and thermo fisher i-port software. These measured values were averaged to attain daily, monthly and seasonal variations. Accuracy of the instruments is sustained by calibrating every fortnight. The measurements techniques, operating principles and calibration methods employed for gas analyzers used for the study are discussed in the following sections.

3. MEASUREMENT TECHNIQUES AND DATA ANALYSIS

The trace gases in ambient air were measured simultaneously for every 5 min interval with Thermo Scientific instruments. These measured values were averaged to attain daily and monthly concentrations. Auto-sampling was made at 10 meters elevation above the ground level. Continuous measurements of O₃ concentration were done by using Model 49i; Thermo Scientific, USA. The operating principle is that, O₃ molecules absorb UV light at a wavelength of 254 nm. The degree to which the UV light absorbed is directly related to the O₃ concentration as

described by the Beer-Lambert law. Lower detection limit of the analyzer is 1 ppbv with the response time of 20 seconds.

The O₃ analyzer is calibrated by the insitu generation of O₃ using O₃ generator. NO_x measurements were done by using Model 42i; Thermo scientific, USA. The analyzer works on the principle that NO and O₃ react to produce a characteristic luminescence with an intensity linearly proportional to the NO concentration. To quantify the NO₂ concentrations it must be transformed into NO before involving in chemiluminescent reaction. NO₂ was converted to NO by molybdenum heated to about 325°C. Lower detectable limit of NO_x analyzer is 0.40 ppbv; with response time 40 seconds. Zero and span calibration of gas analyzers were performed by Thermo scientific multigas calibrator (Model 146i) using NIST traceable standard gas. Data of meteorological parameters such as temperature, relative humidity, wind speed and direction were collected from Automatic Weather Station (AWS).

4. RESULTS AND DISCUSSIONS:

Major rainfall occurs during the southwest monsoon season when the Relative Humidity (RH) is very high. The seasonal variations of the meteorological parameters such as temperature, RH, Solar radiation, and atmospheric pressure are shown in Figure 2 (a-d).

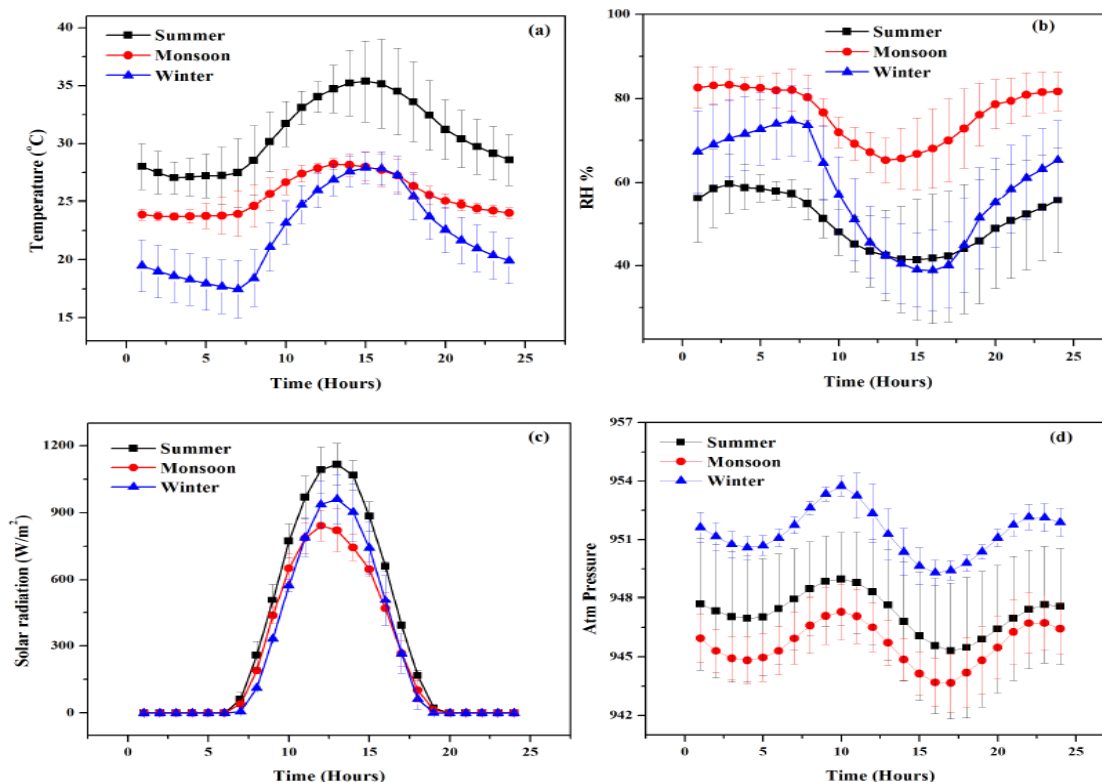


Fig 2: Seasonal-diurnal variations of (a) air temperature, (b) relative humidity, (c) solar radiation, (d) Atmospheric pressure over Hyderabad.

Diurnal profiles of temperature indicated a decrease from evening (04.00 pm) to early morning (06:00 am), and thereafter, increased gradually and reached to a maximum value in the afternoon (2:00–3:00 pm). The increase in air temperature is due to downward solar insolation. Maximum hourly mean temperature ($41.6^{\circ}C$) was recorded the in the month of May during summer season. The monthly mean air temperature is high in April ($32.4^{\circ}C$), while during monsoon it decreases due to increased rainfall and cloudiness. Relative humidity is high at midnight and in the early morning, and it gradually drops after sunrise. The RH exhibits high hourly mean values in monsoon season (88%), followed by winter (86%)

and summer (62%) at night and early mornings. Diurnal profile of Solar Radiation (Figure 2c) shows peak value at mid-day (12:00 pm).

Summer showed the highest mean SR of $1116 Wm^{-2}$, monsoon showed $841 Wm^{-2}$, and winter $960 Wm^{-2}$. The atmospheric pressure is low during monsoon (Figure 2d) suggesting an uplift of moist air masses and release of precipitation. Precipitation is very less and occasionally nearly absent in winter and summer (Figure 3). In contrast, during monsoon (July-October) the rainfall is high reaching a peak. The maximum rainfall was observed in July (about 158 mm). The total rainfall received in monsoon was 370mm, which is 69% of total rainfall in the year.

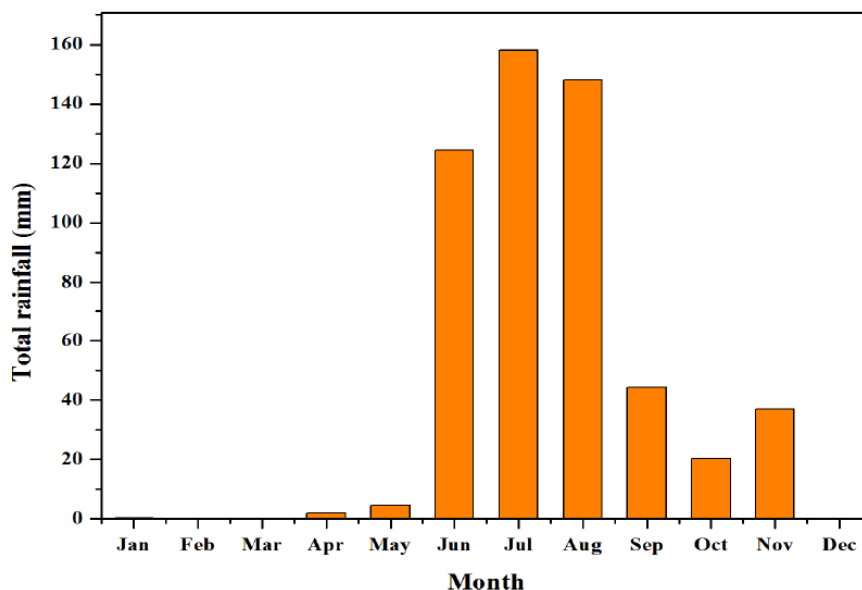


Fig 3: Monthly variations of rainfall over Hyderabad

4.1. Diurnal variations in surface Ozone

Diurnal variations of O_3 during different seasons are presented in Figure 4 for the mean of the years 2010-14. Vertical bars in the figure show the standard deviation (1σ) from the mean. Concentration of O_3 exhibits the typical diurnal pattern, which starts increasing gradually after sunrise, attains maximum value during noontime, and then keeps decreasing gradually in the evening, thereafter maintaining low values over night hours. In the diurnal profile of O_3 , a sudden drop in the morning (07:00–08:00) is attributed to O_3 reduction by titration reaction with NO. A raise in concentration of O_3 was observed after 09:00 am and then reached to maximum at late afternoon hours (12.00pm – 04.00 pm). The high concentrations due to the combined effect of increased atmospheric boundary layer (ABL) height. Low levels of O_3 during nighttime are due to absence of photochemical oxidation and O_3 titration with NO occurs in the residual boundary layer. NOx in the form of NO_2 produces O_3 by photolysis during sunlit hours, and NO in NOx reduce O_3 during nighttime. This confirms the role of NOx in the formation and reduction of O_3 . The variation in night time O_3 values

and photochemical oxidation of the precursor gases such as CO, CH_4 , and VOC in presence of sufficient NOx concentration [1].

Being an urban site, NOx concentrations are commonly observed above the threshold level (~ 10 pptv) in Hyderabad, which is conducive for O_3 production [1]. Since, O_3 concentrations are usually associated with temperature and solar radiation [13, 14]; noontime O_3 concentration was observed to be higher, when temperature and solar radiations attain peak value (Figure 2). Increase in boundary layer height during afternoon is due to convective heating and consequent stratification, leads to the mixing of air in the lower altitude with the air in higher altitude, which is rich in ozone caused increase in concentration of O_3 [15, 16].

are probably due to difference in reactivity of O_3 with anthropogenic components such as NOx, VOCs in different meteorological, and dispersion conditions. Concentrations of O_3 , with larger amplitudes are observed almost throughout the year, except during cloudy and rainy days, due to non-availability of SR and washout of air pollutants, respectively.

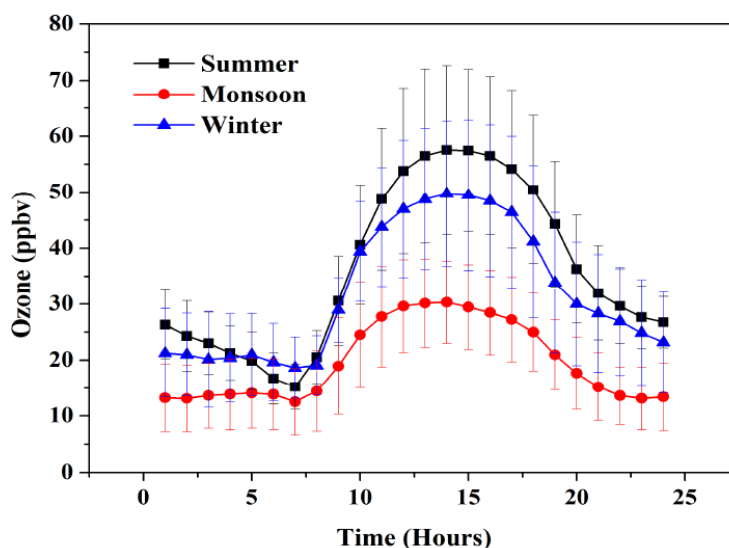


Fig 4: Diurnal variations in ozone during different seasons

4.2. Seasonal variations in surface Ozone

The seasonal variations of O₃ concentrations (Figure 4) revealed apparent and systematic seasonal

variations for a typical geographical location. The increase or decrease in concentration of O₃ could be due to the seasonal variations and related chemical transformations [17].

Table 1: Spatial comparison of O₃ (mean ± SD) at different sites in India

S.No.	Location	Latitude/Longitude	O ₃ (ppbv)	Reference
1	Chennai	13.04°N/80.23°E	15±40	Pulikesi et al. [26]
2	Ahmadabad	23.04°N/72.62°E	24±14	Lal et al. [20]
3	Agra	27.10°N/78.02°E	53±12	Singla et al. [27]
4	Pune	18.54°N/73.81°E	32±13	Beig et al. [28]
5	Kannur	12.26°N/75.39°E	29±50	Nishanth et al. [29]
6	Delhi	28.65°N/77.27°E	24±40	Ghude et al. [30]
7	Kolkata	22.36°N/88.24°E	36±26	Purkait et al. [31]
8	Hyderabad	17.47°N/78.58°E	46±14	Present work

Seasonal O₃ episodes (Figure 4) clearly showed maximum O₃ in summer (57.54 ± 15.07 ppbv) which is attributed to regional photochemistry. In monsoon, the afternoon peak time O₃ level is observed to be low (30.32 ± 7.35 ppbv) due to wet surface deposition of air pollutants by monsoonal rains and scarce availability of solar insulation. In winter, the

peak time O₃ concentration (49.90 ± 12.8 ppbv) is considerably higher than monsoon, attributing to localization of precursor trace gases in shallow boundary layer under humid conditions. Observed mean of O₃ over Hyderabad are compared with other sites in India (Table. 1).

The monthly mean variations of surface and tropospheric column O₃ values are presented in Figure 5. High monthly mean concentration of O₃ was observed in April (36.79 ± 14.38), whereas July (16.73 ± 3.76) and August (14.99 ± 4.71) are associated with low levels of O₃. Sharp increase in O₃ amplitude during October (28.25 ± 8.36) is attributed to the change in the wind pattern from

southwesterly to southeasterly, which brings the trace gases from the surrounding industrially developed areas to the observational site. Tropospheric columnar O₃ also showed similar variations. Low tropospheric O₃ was observed in August (23.4 DU) during monsoon and the highest values in summer (April (40.6 DU)).

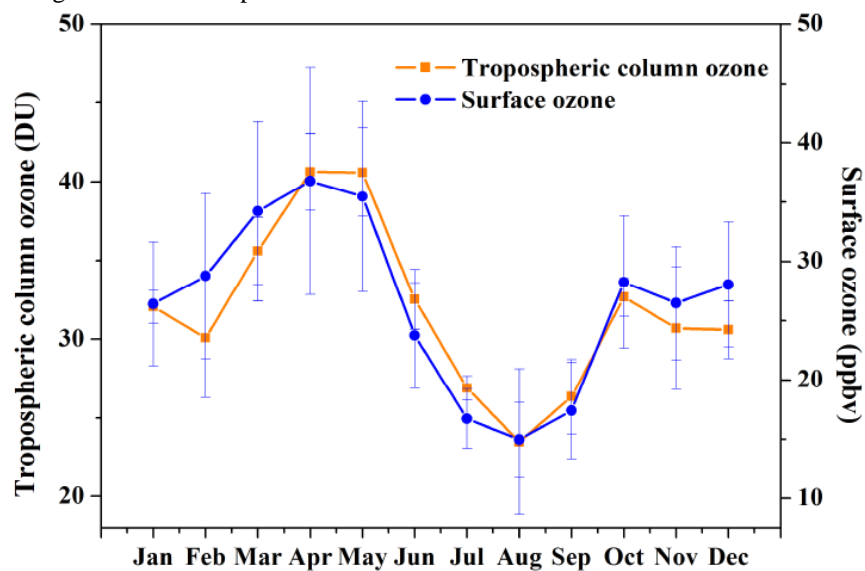


Fig 5: Monthly mean variations in surface and tropospheric column ozone

The wider amplitude of the diurnal cycles during winter and summer is attributed to thermal inversion [20]. These observed changes in O₃ concentration is influenced by several factors in the complex process of atmospheric chemistry, dynamics, and transport of air pollutants. ABL is one such factor in troposphere, where vertical mixing of atmospheric pollutants occurs. Generally, ABL height varies with both time and space.

During daytime, vertical mixing layer of air mass is convectively driven and reached its maximum height by the afternoon due to solar insolation. The convective energy transfer between the surface and air is partly accomplished by turbulent eddies which are produced primarily by wind shear and buoyancy. After sunset, turbulence decays in the mixed layer and a residual layer is formed. During night, temperature decreases throughout the depth of the residual layer causing neutral stratification. Mahalakshmi et al., observed that the ABL height at Hyderabad varies between one to ~3.7 km from

January to December [18]. The ABL was high during summer (March–May) with maximum height in April (~3.7 km), whereas the ABL was shallow in winter (December–February), with minimum height in December (~1.3 km).

4.3. Role of meteorological parameters on Ozone

Most of the atmospheric chemical reactions occur in presence of sunlight. In particular, the shorter wavelength of light with higher energy oxidizes primary pollutants such as NO_x to form secondary air pollutants (O₃). The rate of reaction is dependent on the downward solar flux, which is relative to the latitude–longitude, altitude, time of the day and other radiative factors [19]. Further, chamber experiments also suggested that the immediate NO₂ photolysis when exposed to irradiation resulted in formation of O₃, and the rate of O₃ formation correlates linearly with radiation flux [20]. Therefore, measurement of solar radiation at the study site has significance. The observed surface O₃ concentrations showed a positive correlation with solar flux (Table 2).

Table 2: Pearson correlation matrix of O₃ with different variables

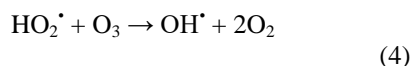
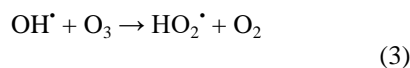
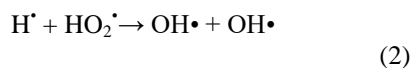
Variables	Summer	Monsoon	Winter
NO	-0.275**	-0.042	-0.221*
NO ₂	-0.353*	-0.260**	-0.461**
CO	-0.241**	-0.237**	-0.438**
SO ₂	-0.115*	-0.066	0.164**
SR	0.517**	0.591**	0.644**
T	0.449**	0.547**	0.503**
RH	-0.082	-0.657**	-0.618**
WS	0.069	0.078*	0.508**
BC	-0.240*	-0.254**	-0.445**

Values with p<0.005 ** Values with p<0.001

The hourly mean noontime solar radiation received during the entire study (Figure 2) showed that summer recorded a maximum value of $1,116 \pm 94 \text{ Wm}^{-2}$, whereas low solar flux was observed in monsoon $841 \pm 68 \text{ Wm}^{-2}$. Ozone showed a positive correlation with temperature (Table 2), which is a measure of incident solar radiation that consequently affects the photochemistry [19, 21, and 22]. Monthly mean variations of solar radiation (Figure 6) showed a maximum in April and minimum mean in the month of July. The high temperature could cause increased O₃ levels during summer months. Summer recorded a maximum mean midday temperature of $35 \pm 3 \text{ }^\circ\text{C}$, while in monsoon and winter, the maximum mean temperatures recorded were 28 ± 2 and $27 \pm 4 \text{ }^\circ\text{C}$, respectively. Monthly mean temperature profile (Figure 6) showed high values in April ($32.4 \pm 4 \text{ }^\circ\text{C}$) and May ($32 \pm 3 \text{ }^\circ\text{C}$) and low values in January ($22 \pm 4 \text{ }^\circ\text{C}$) and December ($20 \pm 4 \text{ }^\circ\text{C}$).

Relative humidity in the atmosphere also played a crucial role in the formation and destruction of O₃

[23]. In low NO_x conditions, hydroxyl radicals can react with ozone leading to further destruction of ozone in a sequence of reactivity chain involving formation of hydroxyl radicals.



Along the diurnal scale, a variation in RH depends on temperature and was inversely correlated with temperature and O₃ (Table 2). Monthly mean RH was highest in July (80.5 ± 7) and lowest in April (40.5 ± 11) (Figure 6).

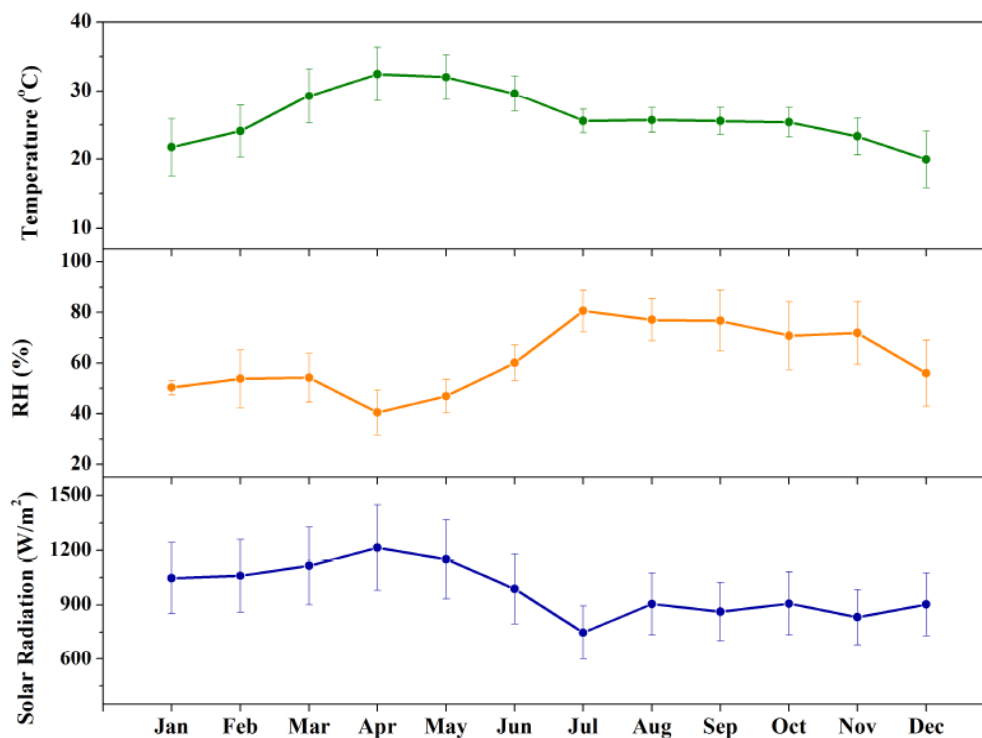


Fig 6: Monthly variations in meteorological parameters (Temperature SR and RH)

On diurnal scale the observed monthly mean maximum and minimum RH were 88.3 % in July during nighttime, and 25.2 % April during afternoon respectively. David and Nair reported that monsoonal air hailing from the marine sources is rich in water vapor content and can cause reduction of O₃ concentration by involving OH[•] radicals [24]. These observations were supported by the air mass back trajectories simulated at the study site.

Wind speed and direction could affect trace gas concentration. Wind speed characterizes mechanical turbulence causing dilution or concentration and transport of air pollutants, while wind direction determines the path of the pollutant dispersion [25, 26]. In monsoon, the wind speed is very strong. However, in winter, wind is very slow and in summer, moderate winds are advent from different directions.

Table 3: Maximum and minimum mean of O₃, NO_x, CO and SO₂ during different seasons

Season	Ozone		NO _x		CO		SO ₂	
	Max	Min	Max	Min	Max	Min	Max	Min
Summer	57.5±15.1	15.2 ±4.1	12.0±2.2	4.0±1.2	410.5±112.1	255.3±78.1	7.1±1.8	3.8±1.0
Monsoon	30.3±7.3	12.5± 3.5	8.5±2.3	3.5±1.1	364.9±66.6	253.1±58.7	5.6±2.1	2.5±0.9
Winter	49.9±12.8	18.6±4.4	15.5±3.5	4.3±0.7	528.2±66.5	313.3±53.1	10.4±3.0	5.6±1.4

Wind speed and direction showed notable changes during February–March, June– July and October November due to seasonal transitions. A fair estimate of dispersion of air pollutants in the atmosphere is possible based on the frequency distribution of wind velocity [27]. Soft winds during summer can influence O₃ concentration at the site. Low concentrations of O₃ were observed in monsoon due to the strong winds (66%) that dilute air by carrying O₃ precursors away from the site. In winter, a combination of calm and soft (80 %) arrived from northeast direction. Therefore, the net O₃ formed is comparatively higher in winter compared to monsoon (Table 3).

5. CONCLUSIONS:

Temporal variations of O₃ and its precursors (NO_x, CO, SO₂) were observed at tropical urban site, Hyderabad, India for the year 2010-14. Observations have been examined with meteorological parameters such as temperature, humidity, solar radiation, and wind speed and direction. Complex atmospheric chemistry, boundary layer dynamics, and local meteorology played a significant role on variations of pollutants. Ozone showed positive correlation with temperature and solar radiation, whereas negative correlation with RH. Along with meteorological conditions, local concentrations of precursor gases also influence the variations in O₃. The surface O₃ concentrations showed a typical diurnal and seasonal pattern, which followed the daily solar and temperature cycle. Diurnal variations of O₃ showed highest concentration during noontime, while lowest concentrations during night and early morning. The high concentrations of O₃ during noontime are due to the combined effect of increased atmospheric boundary layer height as well as photochemical oxidation of the precursor gasses in presence of high temperature and solar radiation. Diurnal variations of NO_x, CO showed a double-peak pattern, one of them observed in the morning and the other at night. These peak concentrations are associated with busy traffic density and entrapping the pollutants in shallow boundary layer. Further, reduction in concentrations during afternoon is due to the increased boundary layer height. Nocturnal chemistry of NO_x played an important role in its potential sequestration and in the formation of radicals that could increase O₃ photochemistry after sunrise. BC along the diurnal scale caused reduction in O₃ concentration and participated in heterogeneous chemistry with NO_x.

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