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# Temporal Evolution and Statistical Behavior of Surface Ozone and Model Observations in Semi-Arid Rural Environment

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## ABSTRACT:

Surface O<sub>3</sub> was carried out using ground based measurement in a semi-arid rural area, Anantapur [14.62 °N, 77.65 °E], Southern India. The rate of increase (morning) was highest in January of about 10.3 ppbv h<sup>-1</sup> and lowest was in June (1.36 ppbv h<sup>-1</sup>), whereas the rate of decrease (evening) was maximum in March (5 ppbv h<sup>-1</sup>) and minimum in June (1.6 ppbv h<sup>-1</sup>). Diurnal annual mean variation of OX (total oxidant) showed maximum concentration was around (17:00 hrs) of about 49.4 ppbv and minimum was on 07:00 hrs (24.6 ppbv). The air chemistry data such as RSN (ratio of sulfur and nitrogen oxides) and OX-NO<sub>x</sub> confirmed that the observational site had a substantial regional impact during winter and summer, which is consistent with the surface wind pattern. Chemical box model (NCAR-MM) was used for simulating the effect of water vapour on surface ozone. Total column ozone effect on surface ozone was analyzed and results were reported.

**KEYWORDS:** surface ozone; rate of change of ozone; total column ozone; Chemical Box Model

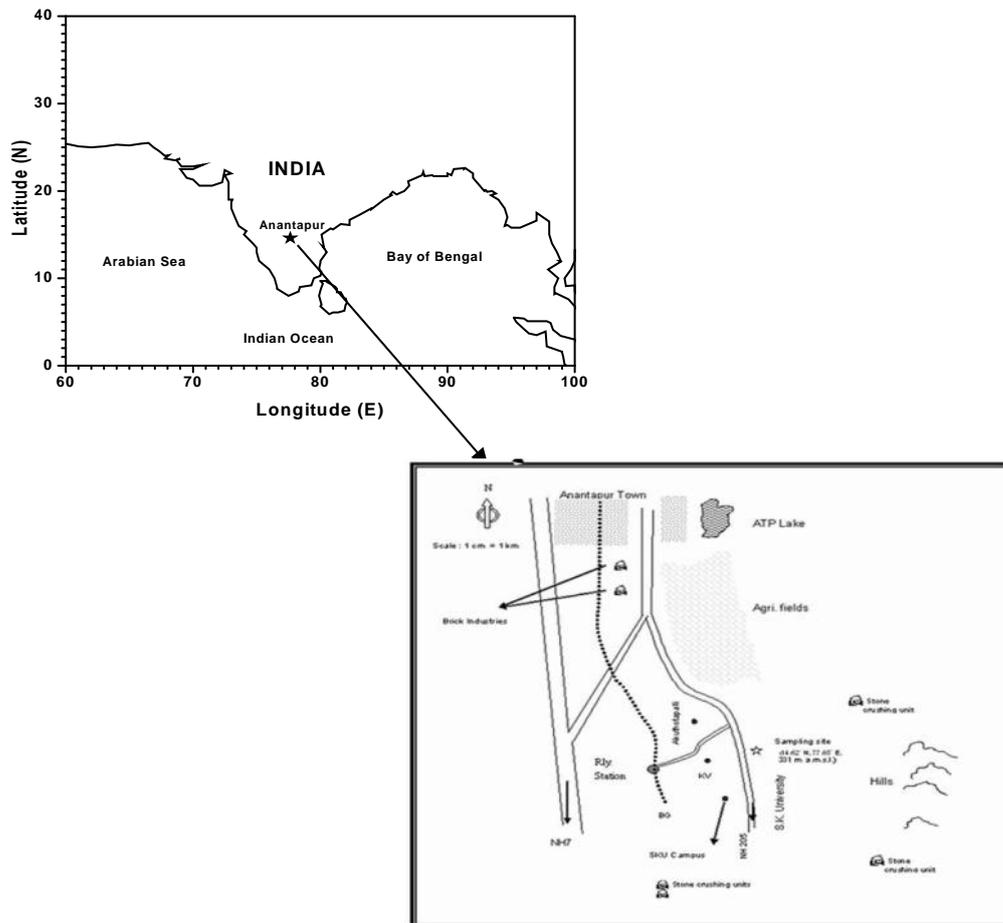
## 1. INTRODUCTION

Surface ozone is a secondary air pollutant, photochemically forms in the atmosphere through a series of complex reactions in the presence of its precursors mainly carbon monoxide, nitrogen oxides, and volatile organic compounds (VOC) (Kleanthous *et al.*, 2014). It plays an important role in oxidation of hydrocarbons, carbon monoxide, dimethyl sulfide (DMS), and sulphur dioxide (SO<sub>2</sub>) (Watanabe *et al.*, 2005). Surface ozone controls a variety of photochemical reactions in the atmosphere as a reactive gas throughout the troposphere (Ravishankara *et al.*, 1998). It plays a central role in the oxidation of chemically and climatically relevant trace gases in the troposphere, thereby regulating their lifetime in the atmosphere. Surface ozone is a major air pollutant that impacts human health, materials, and vegetation (Anenberg *et al.*, 2009). The prevailing winds may carry surface ozone and anthropogenic precursors from emission sources to downwind sites (Sandroni *et al.*, 1994). Therefore, in addition to local emissions, meteorological parameters also play an important role in ambient air pollution. Janjai *et al.* (2016) reported that high surface ozone levels were mainly attributed to biomass burning, solar radiation, and local emissions from motor vehicles over Bangkok, Thailand. The concentration of surface O<sub>3</sub> was influenced by the NO<sub>x</sub>, from motor vehicles and the petrochemical industry in Malaysian Borneo (Latif *et al.*, 2016) and revealed that the meteorological factors, especially temperature, UV radiation, and wind speed, were strongly influenced the surface O<sub>3</sub>. Fiedler *et al.* (2009) revealed that East Asia as the major source region of the pollution and revealed the plume was lifted to the upper troposphere over the Asian continent and transported to Europe across the North America, North Atlantic and North Pacific. Saadat *et al.* (2013) investigated that a considerable amount of air pollutants traveled from beyond the boundary of Bangladesh due to industrial activities in neighbouring India and China. In this paper, the rate of change of surface ozone was discussed. Diurnal variation and local and regional contribution of OX were

reported. Total column ozone effect on surface ozone was studied. Chemical Box model was used to know the effect of water vapor on surface ozone.

## 2. OBSERVATIONAL SITE

The observational site locates in the southern edge of Anantapur, and around 10 km from the main city (Fig. 1). The real time measurements were made at Department of Physics, Sri Krishnadevaraya University, Anantapur. Anantapur is a very dry semi-arid, rain shadow and continental region of Rayalaseema, Andhra Pradesh, India. Maximum rainfall is expected during the monsoon (Southwest monsoon: June–August) and Postmonsoon (northeast monsoon: September–November). The region is surrounded by a number of cement plants, lime kilns, brick making industries and slab polishing with in 50 km radius. The site situated 200 km from Bangalore (urban location) and 350 km from Hyderabad (urban site).



**Fig.1.** Geographic location of Anantapur over the Indian subcontinent and adjacent oceanic regions, also shown road map of the observation site, Sri Krishnadevaraya University (★), Anantapur.

### 3. INSTRUMENTATION

Surface ozone was measured using Model APOA-370, HORIBA, Germany, ambient ozone analyzer. It uses the cross flow modulation type, ultra-violet absorption method in conjunction with the relative calculation method. The optical filter is a band pass filter that passes only light having wavelength of 254 nm. The multipoint calibrator used for calibration and the filter was replaced every four weeks. Total column ozone was derived from OMI Total Ozone Mapping Spectrometer (TOMS). The surface wind flow pattern was obtained from National Center for Environmental Prediction (NCEP) and the National Center for Atmospheric Research (NCAR) reanalysis of monthly data (<http://www.cdc.noaa.gov>).

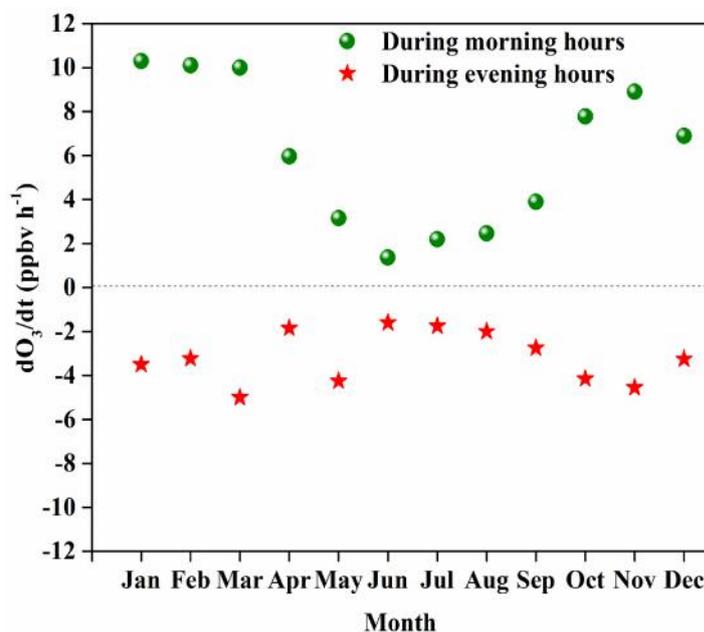
#### CHEMICAL BOX MODEL (NCAR-MM MODEL)

NCAR Master Mechanism was developed at National Center for Atmospheric Research, Boulder, USA. Model was initiated with trace gas species and photon energy is hard wired in the model. The detailed gas phase chemistry of the model has 5000 reactions among the 2000 species and computes the temporal chemical evaluation of an air parcel initialized with known composition. The details of this model were described by Madronich *et al.* (2006).

### 4. RESULTS AND DISCUSSION

#### 4.1. RATE OF CHANGE OF SURFACE OZONE

The rate of change of ozone [ $d(O_3)/dt$ ] during the morning hours (08:00-11:00) and in the evening hours (17:00-19:00) for different months were showed in Fig. 2. The rate of change of ozone during morning and evening times resemble the chemical behavior of surface ozone with its precursors.

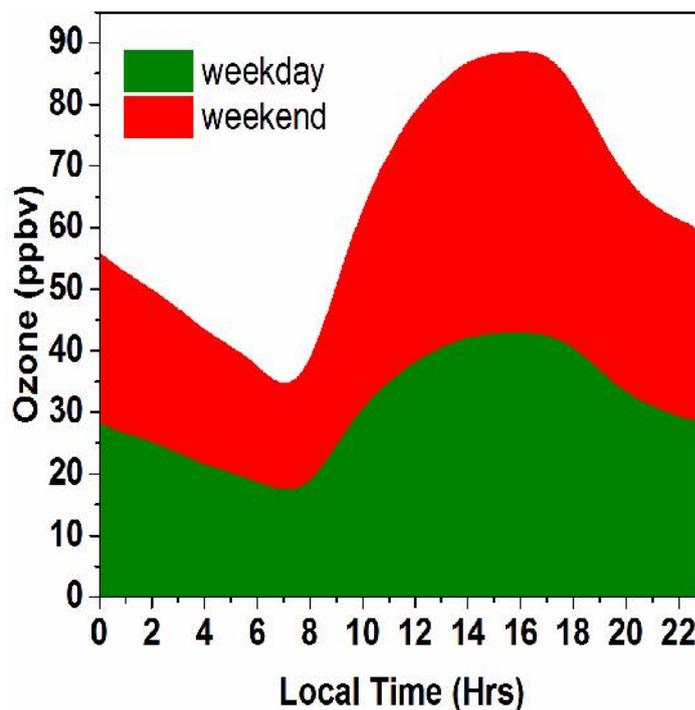


**Fig. 2.** Monthly variation of rate of change of O<sub>3</sub> during morning hours (08:00-11:00) and evening hours (17:00-19:00).

The rate of increase (morning) was highest in January of about  $10.3 \text{ ppbv h}^{-1}$  and lowest was in June ( $1.36 \text{ ppbv h}^{-1}$ ), whereas the rate of decrease (evening) was maximum in March ( $5 \text{ ppbv h}^{-1}$ ) and minimum in June ( $1.6 \text{ ppbv h}^{-1}$ ). The rate of increase was highest in winter ( $9.1 \text{ ppbv h}^{-1}$ ) followed by post monsoon ( $8.3 \text{ ppbv h}^{-1}$ ) and minimum was in monsoon ( $2.48 \text{ ppbv h}^{-1}$ ), while the rate of decrease was maximum during post monsoon ( $4.35 \text{ ppbv h}^{-1}$ ) and minimum during monsoon ( $2.02 \text{ ppbv h}^{-1}$ ). This variation is mainly due to the atmospheric dynamics, photochemical process, and meteorology. In the monsoon sky is generally cloudy, so has less solar flux and substantially reduce the photochemical process. The average  $[d(\text{O}_3)/dt]$  during morning hours was  $6.08 \text{ ppbv h}^{-1}$ , which shows the fast production of ozone by freshly emitted anthropogenic precursors. The low average  $[d(\text{O}_3)/dt]$  during evening hours was observed ( $-3.15 \text{ ppbv h}^{-1}$ ), due to the less anthropogenic activities at the site during these period.

#### 4.2. WEEKDAY AND WEEKEND ANALYSIS OF SURFACE OZONE

The annual mean diurnal variation of weekday and weekend ozone mixing ratios were showed in Fig. 3. We separated the total sampling period into two parts such as weekdays (Monday-Friday) and weekends (Saturday and Sunday). Several studies showed that the surface ozone and its precursor's concentrations are different on weekdays and weekends due to variation in traffic and industrial activities (Qin *et al.*, 2004). The results show that the difference between weekday and weekend ozone concentration was observed to be less and high concentration was observed on weekend than weekday.



**Fig. 3.** Diurnal variations of surface ozone mixing ratios during weekdays and weekends.

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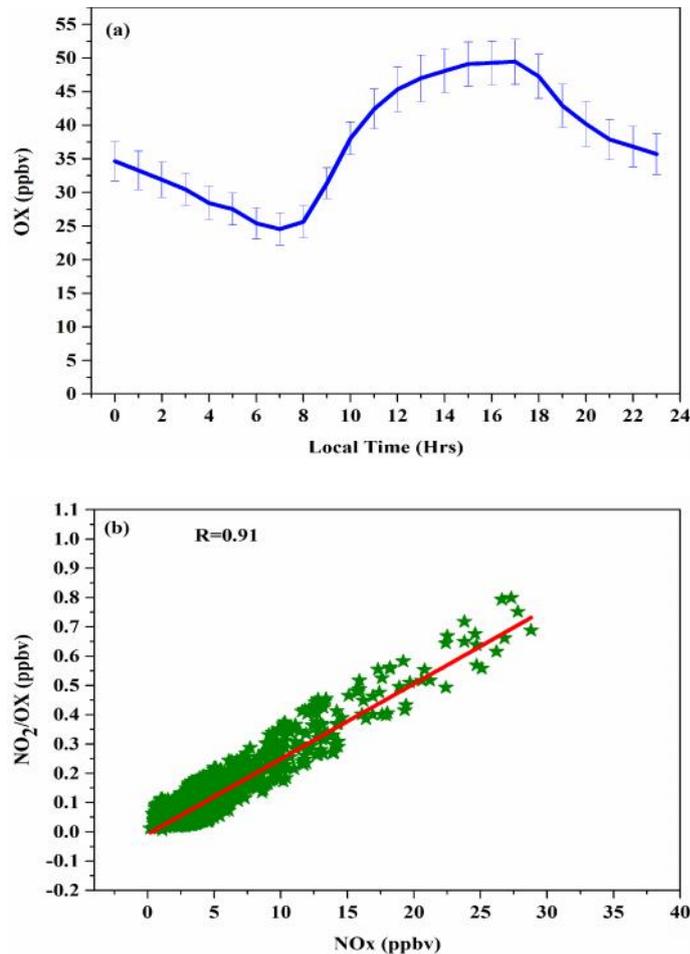
Diurnal profile shows that the maximum concentration was observed on weekdays at around 16:00 hrs (42.8 ppbv) and around 17:00 hrs (45.7 ppbv) on weekends. The 24 hour average ozone concentrations were 30.5 ppbv and 32.2 ppbv for weekdays and weekends, respectively, owing to the titration of ozone with NO that is emitted by anthropogenic sources during weekdays. The measurement location is nearby highway and the number of vehicles will increase during weekdays than weekends and low NO emitted by vehicles and industrial activities on weekends will reduce the capacity of NO titrate ozone (Wang *et al.*, 2014). Sadanaga *et al.* (2008) also reported that ozone mixing ratios were maximum on weekend than weekdays over Tokyo and Osaka, and its precursor concentrations were higher on weekdays. The pattern of higher ozone concentrations on weekends than weekdays were reported in other studies over the globe (Latif *et al.*, 2016; Im *et al.*, 2013; Benas *et al.*, 2013; Sadanaga *et al.*, 2008; Qin *et al.*, 2004).

### 4.3. DIURNAL VARIATION OF OX

The hourly averaged diurnal annual mean variation of OX (total oxidant) was showed in Fig. 4(a). It is defined as the sum of the concentration of oxidation products of species. For OX we adopted only two major components such as O<sub>3</sub> and NO<sub>2</sub>, which account for more than 99% of daytime variation of OX. The total oxidant mixing ratio responds very quickly than ozone mixing ratio for photochemical reactions during daytime (Shiu *et al.*, 2007). Diurnal mean annual variation of OX showed maximum concentration was around (17:00 hrs) of about 49.4 ppbv and minimum was on 07:00 hrs (24.6 ppbv). The OX mixing ratio gradually increase after sunrise and reaches maximum during afternoon and slowly decreases and reaches a minimum during nighttime until the next morning. This variation is mainly attributes to the pollutant loading, photochemical activities, and the height of the mixed layer. The NO<sub>2</sub>/NO ratio suggesting that the high conversion of NO<sub>2</sub> from NO and generates high surface ozone at the site. Diurnal variation of OX exactly coincides with the diurnal variation of surface ozone, suggesting that the variation of OX is less sensitive to NO<sub>x</sub> at the study location.

#### 4.3.1 Correlation between [NO<sub>2</sub>]/[OX] with [NO<sub>x</sub>]

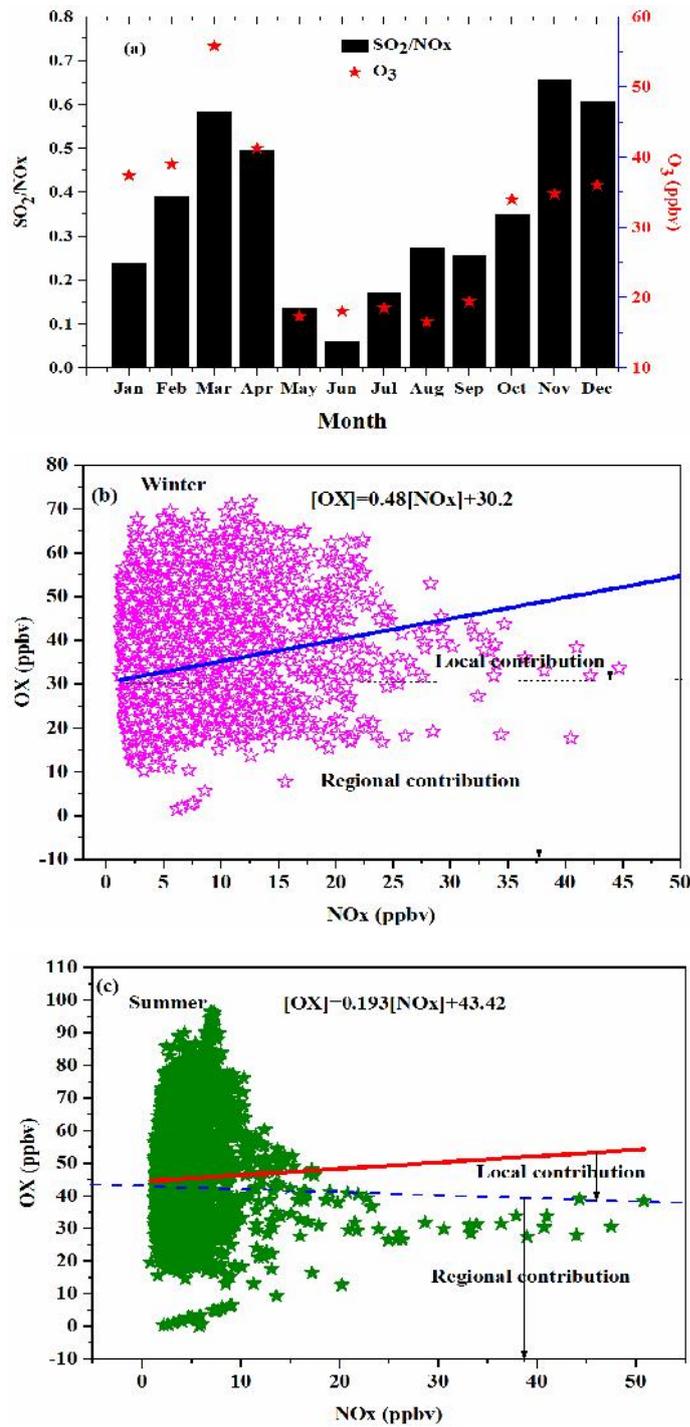
The mixing ratio of total oxidant (OX) is an oxidizing indicator of the O<sub>3</sub> and NO<sub>x</sub>, mainly increases by the background ozone and the conversion of NO<sub>2</sub> from NO. The concentrations of surface ozone and NO were maximum during daytime, while opposite pattern existed for NO<sub>x</sub> and NO<sub>2</sub>, mainly due to the photochemical reactions and mixing process of surface ozone and oxides of nitrogen. The NO<sub>2</sub> forms during nighttime by the reaction of O<sub>3</sub> and NO, which further reacts with O<sub>3</sub> and produces NO<sub>3</sub> and N<sub>2</sub>O<sub>5</sub> and reconvert into O<sub>3</sub> and NO<sub>2</sub> during daytime. The correlation between O<sub>3</sub> and NO<sub>x</sub> was showed negative relation with correlation coefficient (R) of -0.77, attributes to the photochemical reactions in the mixed layer during daytime. The changes of NO<sub>2</sub>/OX with NO<sub>x</sub> mixing ratio was showed in Fig. 4(b), reveals the changes in O<sub>3</sub> mixing ratios with NO<sub>x</sub>. The results suggest that the variations of NO and NO<sub>2</sub> produce linear changes in O<sub>3</sub>, hence NO<sub>2</sub>/OX increased linearly with NO<sub>x</sub>. The data shows that low levels of [NO<sub>2</sub>]/[OX] attributes to the high mixing ratio of surface ozone during noon time and high [NO<sub>2</sub>]/[OX] due to high concentration of NO<sub>2</sub> during nighttime and early morning.



**Fig. 4. (a)** Diurnal variation of OX **(b)** Diurnal variation of  $\text{NO}_2/\text{OX}$  with  $\text{NO}_x$ .

#### 4.4. LOCAL AND REGIONAL CONTRIBUTION

Monthly mean S/N ratio along with surface ozone was showed in Fig. 5(a). An interesting feature was found in this analysis is an obvious increase of  $\text{SO}_2/\text{NO}_x$  associated with the high surface ozone concentration. Monthly mean maximum S/N ratio was about 0.65 in November and minimum in June about 0.05, while the monthly mean maximum and minimum of  $\text{O}_3$  were found in November and June of about 34 ppbv and 18 ppbv, respectively. Diurnal mean seasonal S/N ratio and  $\text{O}_3$  for monsoon, postmonsoon, winter, and summer were about 0.16, 0.32, 0.54, 0.49 and 17, 30, 38, 39 ppbv, respectively. The results suggest that there is an important relation between  $\text{O}_3$  and S/N ratio and the mixing ratio of ozone was strongly depended on regional contribution. It also can be seen that the high levels of ozone observed in winter and summer were associated with the high ratios of  $\text{SO}_2/\text{NO}_x$ , suggesting influence of regional emissions from the surrounding regions at the measurement site.

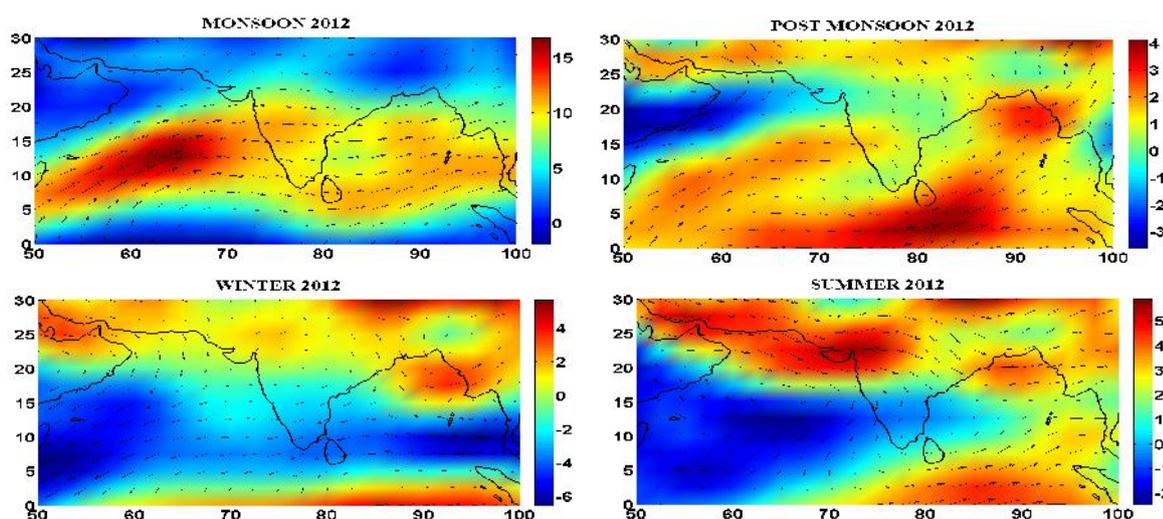


**Fig. 5.** (a) Monthly variation of RSN ( $SO_2/NO_x$ ) and  $O_3$  (b) Variation of  $[OX]$  with level of  $[NO_x]$  during (b) winter (c) summer.

The S/N chemical ratio strongly revealed that noticeable regional contribution to surface ozone at the site during winter and summer. Furthermore, we used another method (OX-NO<sub>x</sub> chemistry) to verify the local or regional contribution to O<sub>3</sub> during winter and summer. The variation in OX concentration with the level of NO<sub>x</sub> for winter and summer was showed in Fig. 5 (b) and (c). In this study, the concentration of OX (O<sub>3</sub>+NO<sub>2</sub>) can be described in terms of the sum of a NO<sub>x</sub>-independent 'regional' contribution (O<sub>3</sub> background), and NO<sub>x</sub>-dependent 'local' contribution (Jenkin, 2004). The slopes (intercepts) for OX vs. NO<sub>x</sub> were 0.48 (30.2) and 0.19(43.4) for winter and summer, respectively. The slope obtained from the regression analysis represents the local OX contribution, while the intercept represents the NO<sub>x</sub>-independent regional contribution. The most important local NO<sub>x</sub>-dependent contributions are traffic exhaust and oxidant coming from NO reaction with oxygen to form NO<sub>2</sub>, which depends both on the NO concentration and photolysis rate of radical formation. The positive correlation between NO<sub>x</sub> and OX provided at the site, showing clear evidence that the local sources substantially contributes in the formation of ozone. Furthermore, it can be seen that the OX at a given location had a more NO<sub>x</sub>-independent contribution (high intercept) (Fig. 5 b and c). The above analysis also suggested that significant regional contribution was found for ozone formation during winter and summer at the study site.

#### 4.5. NCEP/NCAR REANALYSIS

Fig. 6 shows the synoptic pattern of monthly mean surface wind speeds for different seasons over the Indian subcontinent, with the arrows head showing wind directions and the lengths of the arrows representing the wind speeds (m s<sup>-1</sup>). The results show that strongest winds prevailing from southwest of Arabian Sea during monsoon. This scientific study of wind pattern effectively confirms that domination of local contribution over the site during monsoon, which is consistent with the air chemistry data analysis. During winter, winds were predominantly from the east or north east and were from the polluted northern regions surrounding India. These regions were reported to be the potential source for pollutants. The winds with high speed were originated from W/NW direction and from regional pollutant areas during summer, confirm that regional contribution over the site during these periods. The air chemistry data such as RSN and OX-NO<sub>x</sub> confirmed that the observational site had a substantial regional impact during winter and summer, which is consistent with the surface wind pattern.

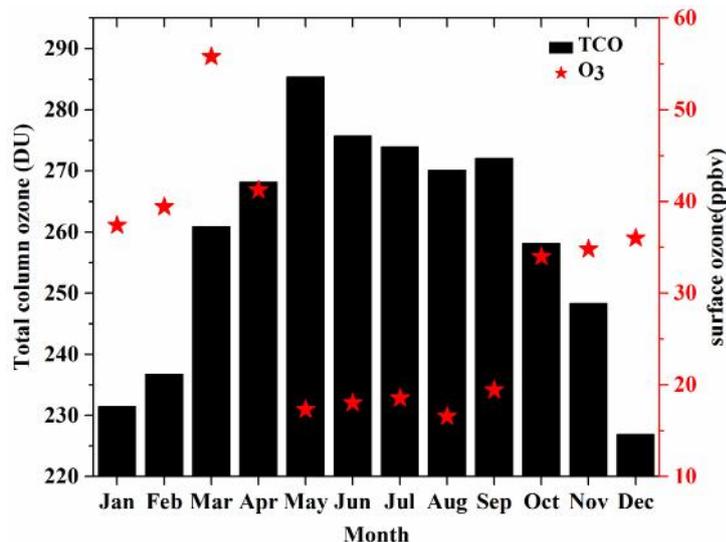


**Fig. 6.** Mean prevailing winds at 850 hpa over Indian region during monsoon, postmonsoon, winter and summer (from NCEP/NCAR reanalysis data).

## SATELLITE OBSERVATIONS:

### 4.6. TOTAL COLUMN OZONE EFFECT ON SURFACE OZONE

Monthly mean variation of total ozone and surface ozone was showed in Fig. 7. The data collected by the Ozone Monitoring Instrument (OMI) and the Microwave Limb Sounder (MLS). The total ozone increased from March and reached maximum in May, remained high until October and decreased in November onwards to till February. The maximum total ozone was observed in May around 285 DU and minimum of about 226 DU in December. For seasonal variation maximum total ozone was observed during monsoon around 272 DU, followed by summer of about 271 DU and minimum was found in winter around 231 DU. This variation is mainly attributes to balance between transports associated with the diabatic mean circulation of the stratosphere, strong photochemical activity, and transport from the lower troposphere into the upper troposphere and lower stratosphere (Antón *et al.*, 2011). Similar monthly variation of total column ozone was observed over Mexico, the low values were occurred during December and January, while high values between April and May (Zhang *et al.*, 2015).



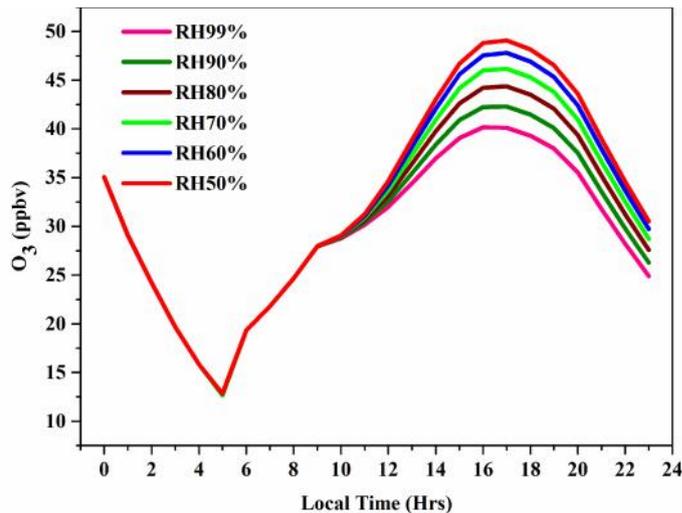
An interesting **Fig.7.** Monthly variation of total column ozone with surface ozone. It is that the enhanced peak in surface ozone was found that the strong negative relation between total ozone and surface ozone. The effect of total ozone on surface ozone is mainly due to the high absorption of ultraviolet radiation around 300 nm that would otherwise reach the surface. The surface ozone formation is strongly depends on the ultraviolet radiation around 300-320 nm.

## MODEL RESULTS:

### 4.7. EFFECT OF WATER VAPOR ON SURFACE OZONE

In the lower atmosphere water vapor is actively participated in atmospheric chemical reactions and it is powerful oxidizing agent in the atmosphere. Diurnal variation of ozone was simulated for different RH (Relative Humidity) values to understand the effect of water vapor on surface ozone concentration. The model simulates chemical evolution of an air parcel initialized with known concentrations of various species. The observed diurnal variation of meteorological parameters, latitude, longitude, column O<sub>3</sub>, NO<sub>2</sub>, aerosols, and boundary layer height were used in the model simulation. The background concentrations of methane and non methyl hydrocarbons, CO, NO<sub>2</sub>, and O<sub>3</sub> were also used. The model was simulated with same gas phase chemistry and changes in RH from 50% to 99% to understand the water vapor effect on surface ozone. The

model simulation resulted that diurnal variation of ozone was decreased with increased water vapor (Fig. 8). The surface ozone was decreased by 3.2% at 50% of RH, and the decreasing rate was increased to 12.4% at 99% of RH. This is mainly due to the Photolysis of ozone produces  $O(^1D)$  that reacts with water vapor, produces OH radicals and results ozone depletion (Pitts and Pitts, 2000). We concluded by this model analysis higher water vapor content in the lower atmosphere had played a substantial role in ozone destruction at the measurement site.



**Fig.8.** Model simulation of change in ozone with increase of Relative Humidity (RH).

## CONCLUSIONS

The average  $[d(O_3)/dt]$  during morning hours was  $6.08 \text{ ppbv h}^{-1}$ , which shows the fast production of ozone by freshly emitted anthropogenic precursors. The results suggested that the variations of  $NO$  and  $NO_2$  produce linear changes in  $O_3$ , hence  $NO_2/OX$  increased linearly with  $NO_x$ . The air chemistry data such as  $RSN$ , and  $OX-NO_x$  confirmed that the observational site had a substantial regional impact during winter and summer, which is consistent with the surface wind pattern. The results showed that the difference between weekday and weekend ozone concentration was observed to be less and high concentration was observed on weekend than weekday. Maximum total ozone was observed during monsoon around 272 DU, followed by summer of about 271 DU, and minimum was found in winter around 231 DU. The surface ozone was decreased by 3.2% at 50% of RH, and the decreasing rate was increased to 12.4% at 99% of RH and concluded by this model analysis higher water vapor content in the lower atmosphere played a significant role in ozone destruction.

## ACKNOWLEDGEMENTS

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