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Advances in Applied Science Research, 2013, 4(5):309-320

ISSN: 0976-8610
CODEN (USA): AASRFC

# Diurnal and seasonal characteristics of ozone and NOx over a high altitude Western Ghats location in Southern India

**C. Udayasoorian[1]*, R. M. Jayabalakrishnan[1], A. R. Suguna[1], S. Venkataramani[2] and Shyam Lal[2]**

[1]*Department of Environmental Sciences, Tamil Nadu Agricultural University, Coimbatore, Tamil Nadu, India*
[2]*Physical Research Laboratory, Ahmedabad, India*

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**ABSTRACT**

*Continuous measurements of ozone and NOx concentrations were made over a period of two years from a high altitude location Ooty in the Nilgiris Mountain Range in southern India to characterize the diurnal and seasonal variations. The ozone and NOx concentrations were higher during warmer months and prominent seasonal variations were recorded. A seasonal variation in ozone showed a pronounced maximum (62 ppb in March) in summer with values sometime exceeding 90 ppb and a minimum in the monsoon and post monsoon season (17 ppb in August). Interestingly, diurnal pattern of ozone did not show any day time build up throughout the study period. On the contrary, lower ozone mixing ratio was observed during the day. A monthly maximum NOx value was observed during summer month of April (1.85 ppb), whereas minimum value was observed during monsoon (0.19 ppb in August). Further, low rainfall, high temperature and solar radiation have given rise to the gradual build up of ozone during summer and winter. The unique meteorology over this region seems to play an important role in seasonal and as well as diurnal variation of ozone. Regional pollution is shown to have maximum contribution to ozone level during February to May.*

**Keywords:** Ozone, Oxides of nitrogen, High altitude site, Correlation.

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## INTRODUCTION

Trace gases in the atmosphere present a major impact on the environment and troposphere ozone ($O_3$) is considered to be the most widespread air pollutant and important radiatively active trace gas [32, 43], by indirect greenhouse effect, associated with the emissions of ozone precursor species because of their potential impact on the trophospheric ozone distribution [10].The ozone content in the troposphere is substantially influenced by human activity, atmospheric emissions of nitrogen oxides (NOx), carbon oxides (CO and $CO_2$), methane ($CH_4$), respirable suspended particulates (RSP), sulphur dioxide ($SO_2$) and volatile organic compounds (VOC), promote ozone formation in polluted air and increase the ozone concentration in the troposphere [47]. NOx and VOCs are emitted from anthropogenic sources such as fossil fuel power plants, industrial activities and transportation, as well as natural sources such as lightning, soil (NOx), and vegetation (biogenic VOCs such as isoprene) [42]. High concentrations of ozone are considered as a problem with serious repercussions on living organisms [2] and the critical level is known to be a toxic for human and phytotoxic to vegetation [43,46]. However, in the past decades, anthropogenic activities have led to increase in the level of trophospheric ozone beyond the critical limit throughout the world [32, 45]. Besides the surface ozone concentration is, mainly controlled by local and regional emissions [44, 11], though they could also be affected by large scale transport. In this context, study of spatial, temporal distribution and characteristics of ozone and NOx is crucial in the understanding of trophospheric chemistry over different geographic locations assumes importance. The temporal variations of $O_3$ have been reported at many sites including rural, urban, coast and mountain sites in India [16, 21, 9, 23,]. These studies also showed great spatial variability. For instance, higher ozone values did not appear during daytime at a mountain site in India [23], while significant increasing ozone values during daytime were observed at an urban site, Ahmedabad, in tropical India [16]. However large gap areas still exists geographically. In view of the above, continuous measurement of ozone

and NOx was initiated from Ooty, a high altitude location in Western Ghats of southern India in the Nilgiris Mountain range, to understand the ozone characteristics over this remote environment; and the changes in the precursors associated with dominant source types and consequences of regional weather and climate implications. In this paper, we present the results of ozone and NOx measurements carried out for the period from March 2010 to March 2012 under ISRO-GBP – ATCTM. (Indian Space Research Organisation – Gio - Biosphere Programme, Atmospheric Trace Gases – Chemistry Transport and modelling project)

## MATERIALS AND METHODS

### EXPERIMENTAL SITE AND DATABASE

The study area, Ooty (11.4 °N, 76.7 °E, 2520 m amsl; Fig.1), located in the district of the Nilgiris Mountain range in Indian state of Tamil Nadu., represents a fresh, clean and pleasant environment with highly subdued human activity. Dense forests, lofty mountains, extensive tea and coffee plantation and sprawling grasslands characterize the location. The Nilgiris Hills form a part of a larger chain of mountains known as the Western Ghats along the western side of India, which is one of the eight hottest hotspots of biological diversity in the world. As of 2001 India census, Ooty had a population of 93,921.

The continuous observation of surface ozone have been made since February 2010 using ozone analyzer (Thermo Fisher U.S.A Model 49*i*) which is based on the well known technique of UV absorption. The analyzer aspirates air from a height of about 5 m above the ground level through Teflon tube with a flow rate of about 1 L per minute. The minimum detection limit of the analyzer is about 1 ppb. Calibration is done at frequent intervals using zero air and known span values.

The continuous measurement of NOx have been made using NOx analyzer with Chemiluminescence technology, the Model 42i-TL measures the amount of nitrogen oxides in the air from sub-ppb levels up to 1000ppb. The Model 42i-TL is a single chamber, single photomultiplier tube design that cycle between the NO, NOx and Zero modes. The addition of the Zero mode provides for excellent long term stability and extremely low minimum detectable limits. The minimum detection limit of the analyzer is about 50pptv. The calibration of the system was done using a reference standard $NO_2$ cylinder.


**Figure 1. Geographical position of Ooty in Western Ghats**

CNR 1 net radiometer is used to measure the solar radiation and Far Infrared radiation separately. Solar radiation is measured by two pyranometers, one for measuring incoming radiation from the sky, and the other, which faces downward, for measuring the reflected solar radiation. From these two pyranometers, albedo, the ratio of reflected and incoming radiation, can also be determined. The other meteorological data obtained from the automatic weather station, Horticultural Research Station, Tamil Nadu Agricultural University, Ooty.

**GENERAL METEOROLOGY**

The general meteorological conditions that prevailed over Ooty are characteristics of a subtropical highland climate. Despite its location in the tropics, Ooty generally features pleasantly mild conditions throughout the year in sharp contrast with most of South India. However, night time in the months of January and February is typically chilly. The monthly mean values of Temperatures are relatively consistent throughout the year; with average maximum temperatures ranging from about 17-20 degrees Celsius and average minimum temperatures between approximately 5-12 degrees Celsius. The average precipitation is 1250 mm annually, with a marked drier season from December through March. The temporal variations of meteorological parameters (Temperature, Relative Humidity and Rainfall) during the measurement period are shown in Fig.2 while that of wind speed and wind direction are shown in Fig.3. As can be seen from Figure.2, the monthly temperature reached maximum value (~ 20°C - 25°C) during the summer (March to May) season and a minimum (~ 15°C – 17°C) during monsoon (June to August) season. The relative humidity was generally high (> 50%) throughout the period of observation. Seasonally, averaged rainfall is higher during monsoon and post monsoon (September and November) season which accounts 80% of total rainfall.

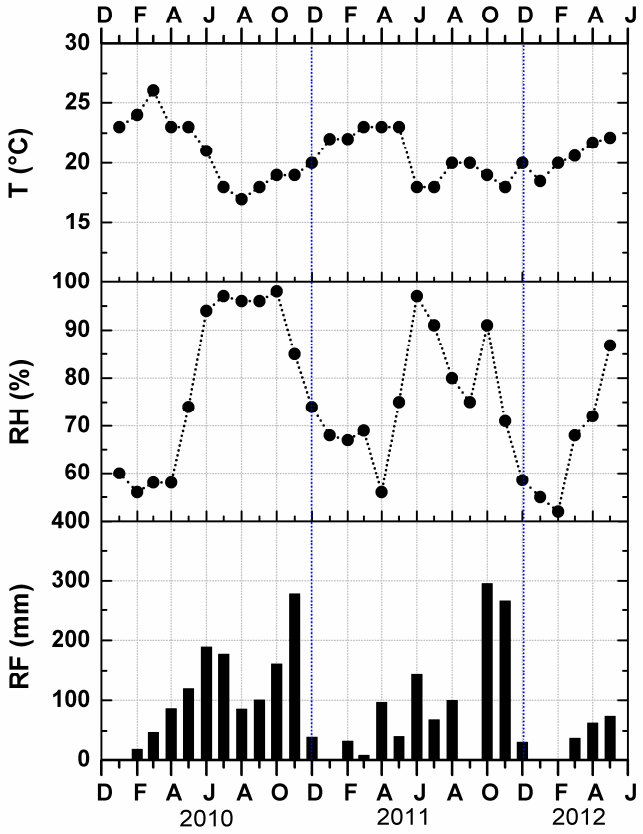


**Figure 2. Temporal variation of monthly mean meteorological parameters (T, RH, RF)**

The winds however are highly seasonal, with low and moderate (< 4 $ms^{-1}$) northeasterlies/ easterlies dominating the winter (December to February) and summer, changing over to westerlies in monsoon and autumn season as shown by the polar diagram in Fig.3.

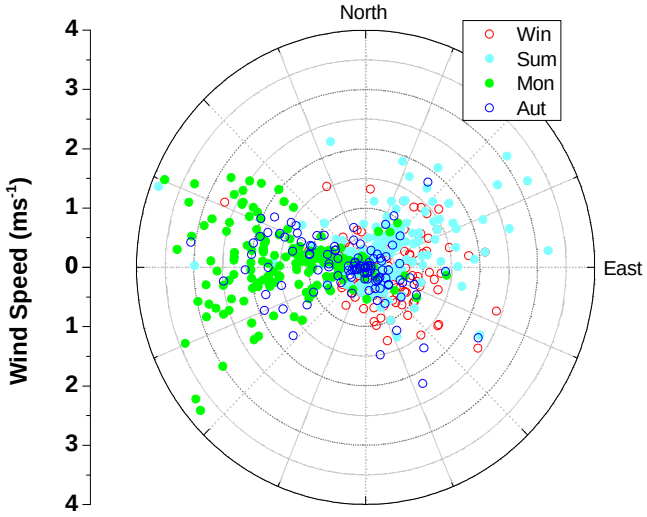


**Figure 3. Wind speed and wind direction at different seasons**

## RESULTS AND DISCUSSION

***Diurnal variation of ozone concentration***

The diurnal variation of ozone depends on the altitude, solar zenith angle, advection processes, temperature, and the concentrations of halogens such as NOx, HOx, ClOx (13). The 15 minutes average of diurnal variation of ozone in different months representing four seasons *Viz.*, summer (MAM), monsoon (JJA), post monsoon (SON) and winter (DJF) shown in fig.4. The diurnal pattern of ozone was influence by the absence of day time buildup throughout the year. The pattern characterized by high ozone level during night time while, low level during day time. Average ozone values show a decrease immediately after sunrise that is followed by a systematic increase in the evening and thus lower mixing ratios are common during daytime at the site. The same diurnal pattern of ozone was observed at clean / high altitude sites of Mauna loa, 19.5°N, 155.6°W, 3397 m amsl, Mt. Fuji, 35.35°N, 138.73°E, 3776 m amsl and Mt. Abu, 24.6°N, 72.7°E,1680 m amsl, Nainital 29.37°N, 79.45°E, 1958 m amsl, [26, 46,22, 15]. The lower level of ozone precursor (NOx) is anticipated for the absence of day time build up in ozone diurnal variations and low NOx regimes and the prevailing photochemistry leads to net destruction of ozone at noon hours in Ooty. [15,28]. Generally in higher altitudes the mountain valley induced system and chemical ozone loss in low NO environments and solar heating which warms the air near the mountain surfaces and it causes warm upslope wind after the sunrise. This induces the ozone loss by surface deposition during noon time [16] and the levels of precursor gas such as NOx also anticipated being lower at Ooty. Due to surface dry deposition could be characterized by lower ozone concentrations with respect to precursors more representative of the free troposphere. On the other hand, the day time ozone minimum may also be due to ozone loss by surface deposition along the valley floor or *in situ* photochemical destruction under low NOx conditions which are likely to take place at mountain sites [5]. At night, above the nocturnal boundary layer do not experience the same depletion due to absence of mixing precursors for ozone formation. Senik and Elansky [37] reported that after sunrise in the free troposphere at low NOx concentrations and intense UV illumination, the photochemical processes result in the decrease of ozone at noon time and the low NO concentration can lead to a day time surface ozone concentration decrease. Moreover, ozone can be considered as reservoir species and the concentration increases with altitude and the model calculations showed that the concentration of gaseous nitric oxides decreases with altitude and also the volume of ozone mixing ratio increase above 80 Km at night time [33]. Hence, the absence of daytime buildup in diurnal variations of ozone with moderate to lower levels of precursor together suggest that the present site is reasonably away from emission sources and local photochemistry contributes to ozone variations at Ooty.

Diurnal variations in ozone showed a different pattern during summer and winter and less pronounced during June-November is noticeable. Change in the wind patterns in different seasons and the changes in boundary layer mixing height in day and night are the primary factors which are responsible for the seasonal change in the diurnal patterns of ozone [23]. Such behaviours suggest an important influence of thermal winds in flushing out valley bound pollutants to the experimental location during summer leading to highest ozone concentration. With the advent of monsoon, diurnal variation is less pronounced as the much shallower boundary layer isolates the measurement site from the surrounding valley region and washout of the pollution during rainy season.

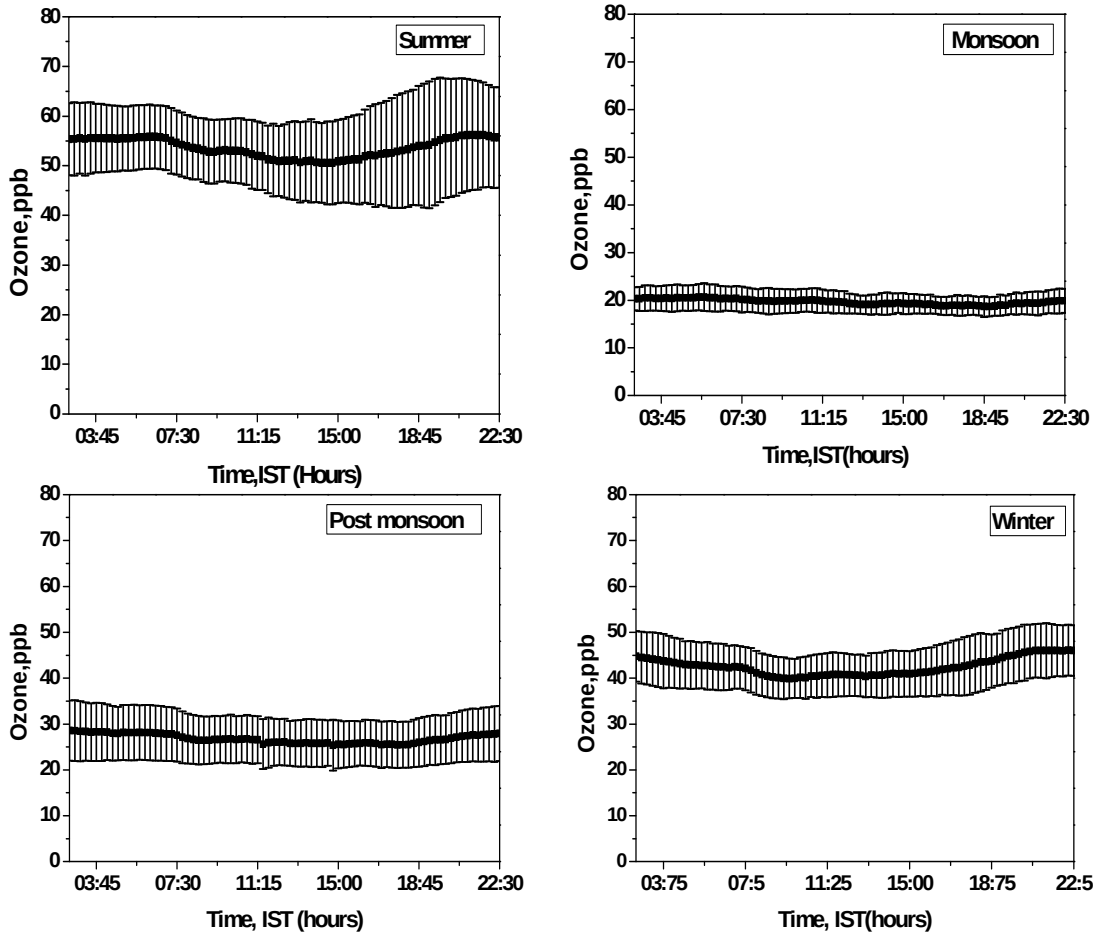


**Figure 4. Diurnal variation of Ozone mass concentration during February 2010 to March 2012**
*(Vertical bars through the measurement points are standard deviation of the mean)*

***Frequency distribution of ozone***

The frequency distribution of daily mean ozone at different seasons showed in Fig.5. During summer, more than 90% of ozone values remain above the annual mean; while most of the ozone values remain below the annual mean during monsoon. The higher ozone abundance during the summer is attributed to the increased vertical transport of effluents in the upwind valley regions, which might have been confined to the valleys within the very shallow boundary layer. The extended duration of daytime and abundant availability of solar radiation, low relative humidity, rainfall, wind speed and wind source from eastern locations during Jan to May sets in thermal convections over these regions leading to the higher mixing concentration of ozone. In addition, the upslope thermal winds which brings ozone rich air from aloft to higher ozone concentration at free trophospheric conditions or higher altitude site. During monsoon, as the temperature decrease to minimum, the atmosphere becomes quite stable, and the ABL shallower, thus enveloping the valley region and isolating the measurement site (peak) from local and regional emission. During the monsoon washout, as well as the very reduced ABL (due to low solar elevation) lead to lower value of ozone, which gradually increases towards winter mainly due to the increase in local anthropogenic activities in the inhabited areas of the Ooty town. With the advent of summer, the increased convection leads to the deepening, eventually breaking up of the surface layer as the season advances, flushing up particles to be lofted and dispersed spatially by the prevailing winds. Finally the low concentrations of NOx, precipitation, and air temperatures influence ozone concentrations throughout the troposphere during monsoon. This would account for initial sharp increase in ozone concentration at the peak during summer (e.g., March).

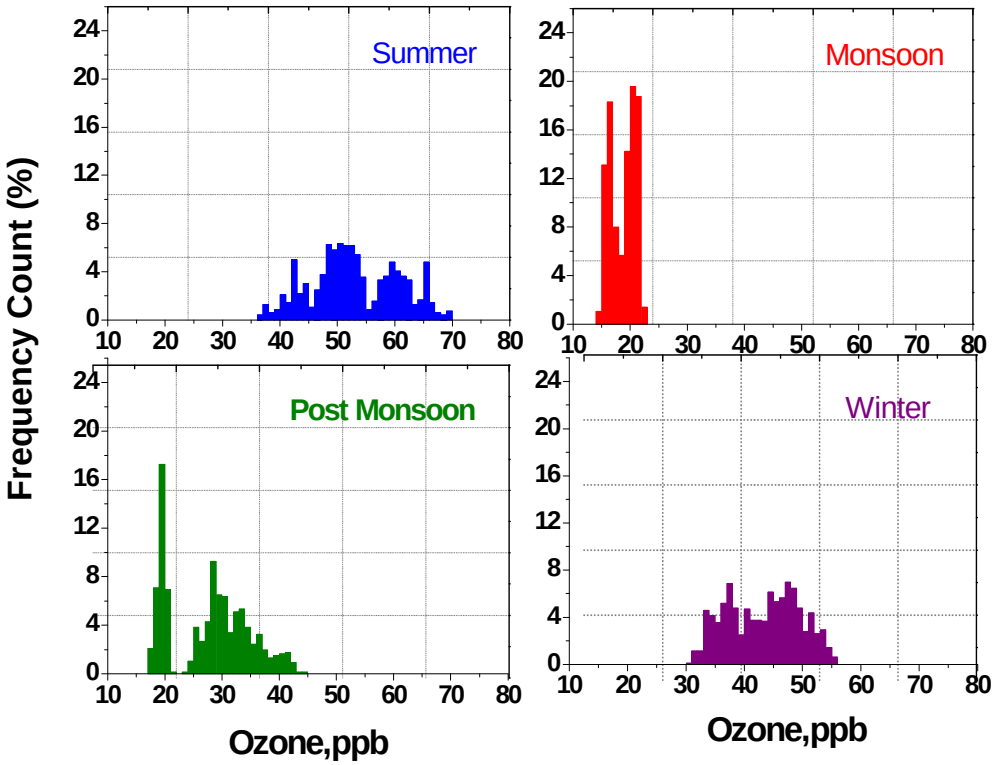


**Figure 5. Frequency distribution of ozone at different seasons**

***Annual and seasonal Variation of Ozone***

The shape and amplitude of ozone cycles are strongly influenced by meteorological conditions (temperature, solar radiation) and prevailing levels of precursors (NOx). The seasonal pattern of ozone is caused by the variation in transport as well as the varying source and sink strengths play a major role in determining the surface ozone variations. The maximum ozone concentration was observed during the month of March 2010 (62 ppb) and minimum ozone value of 17 ppb was observed during August 2010 (Fig.6). The climatological mean estimated from the entire data (irrespective of years) has been 36 ppb. The seasonal mean values (ppb) of ozone for the 2 years period are 53.45 ± 8.18, 19.81 ± 2.4, 27.2 ± 5.67 and 42.81 ± 5.65, respectively for summer, monsoon, post monsoon and winter. The seasonal maximum of ozone concentration recorded during summer and falls rapidly as monsoon approaches. A systematic increase in ozone is observed from March to May (53.45 ± 8.18) thereby it showed a dramatic decrease with lower mixing ratio continuing till August (19.81 ± 2 in monsoon). The secondary maximum of ozone is also observed during Dec. to Feb. Similar, seasonal variation was found at other high altitude regions (Darjeeling), it shows a large seasonal variation from 20 ppb (July- August- September) to 70 ppb (March-April-May) [12]. Generally, ozone levels are higher at high altitude rural sites [29] and ozone is observed to be higher in spring and summer at many measurement sites over the globe. Some high altitude sites (e.g., Mt. Fuji, 35.35°N, 138.73°E, 3776m amsl and Mauna Loa, 19.5°N, 155.6°W, 3397 m amsl) show a springtime maximum value which is largely explained on the basis of downward transport of ozone rich air from higher altitudes [42, 27]. In contrast to the remote sites, there are a number of surface sites that show a broad summer maximum in ozone concentration. The existence of a broad summer maximum is often associated with the photochemical production of ozone [19, 10, 7, 20]. . Many of the higher altitude sites are continental in nature and influenced by pollution [20, 36].The seasonal minimum mixing ratio of 19 and 27 ppb was observed in monsoon and post monsoon period, respectively. Naja *et al*. [23] also reported that large variability of ozone found in Mt.Abu. During monsoon period and seasonal variations of ozone in different climatic zones are largely affected by rain [18]. Increase in ozone mixing ratio after the monsoon season is clearly visible at this site. The mixing ratio of ozone closely linked with the monsoon period. The southwest monsoon starts during June and gets weakened by August, whereby northwest monsoon prevails from September to November at this region.

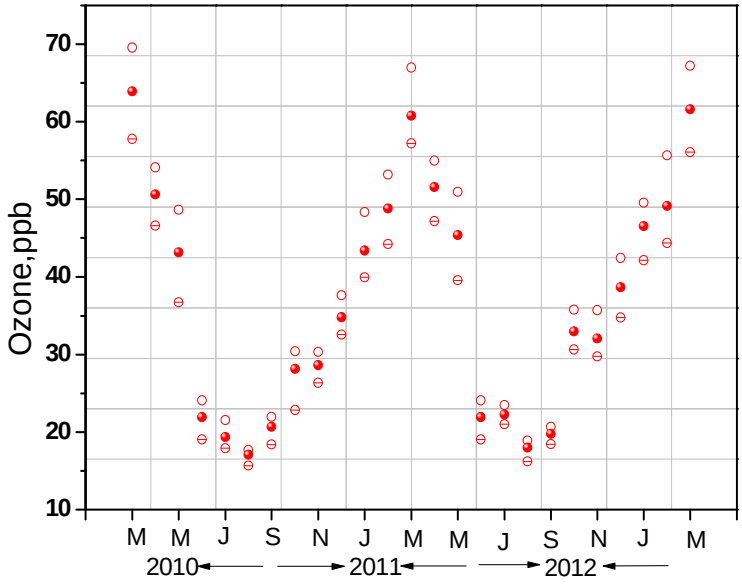


**Figure 6. Temporal Variation of Ozone**

During monsoon period the sky is generally overcast, decreasing the solar insulation and thereby reducing the photochemical processes and pollutants get washed out. These two possible factors could be the cause for low level of ozone recorded during monsoon period.

***Role of long range transport***

With a view to understand the influence of long-range transport of pollutants in determining the ozone characteristics at Ooty, we have examined the 5-day isentropic HYSPLIT back-trajectory analysis, ending at 500 m AGL, as shown in Fig.7 for two reprehensive seasons of summer and monsoon. Consistent with the local meteorological data, the synoptic wind also shows distinct advection pathways with dominant south-westerly advection during monsoon, but strong easterly contribution during summer in addition to the north-westerly airmass component. Thus it appears that the potential sources were mainly distributed to the eastern locations of Ooty, i.e., anthropogenically polluted metropolitan cities like Chennai, Bangalore etc, which contributes mostly to the highest ozone concentration during summer.

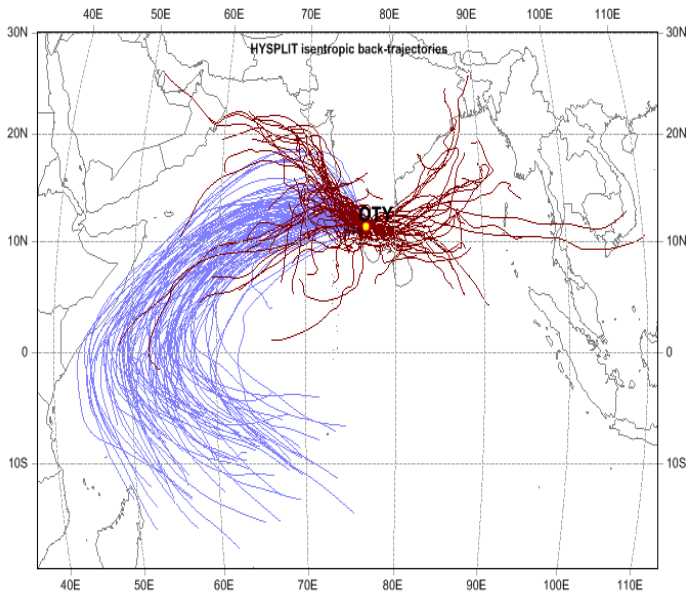


**Figure 7. HYSPLIT isentropic back-trajectories during summer (maroon) and monsoon (blue) seasons**

***Diurnal and Seasonal Variation of NOx***

NOx plays a critical role in the photochemical formation of ozone and has been found to be a limiting factor in the atmosphere at rural and remote locations. On the other hand, in many urban areas of the world air pollution is mostly characterised by the formation of ozone and other oxidants. It may be assessed that in polluted regions, primary pollutants are NOx and volatile organic compounds (VOC) mainly produced by mobile sources, which undergo photochemical reactions in sunlight to form a host of secondary products, the most prominent of which is ozone [4]. Diurnal variation of NOx (Fig.8) showed build up during morning and late evening hours, which are contradictory to the variations of ozone observed, on the other hand winter and summer months recorded higher levels of NOx, which might be due to the anthropogenic emissions. The diurnal variation of NOx concentrations shows a potential and direct link with the ozone variability indeed, at night time, where anthropogenic activities are quite nonexistent. During this time, NOx concentrations show a net decrease and in parallel, we observe a night time ozone increase. In the early morning, a NOx concentration shown a sharp increase and reaches a maximum due to anthropogenic emissions. Then, the first titration reaction of $O_3 + NO \rightarrow NO_2 + O_2$ leads on one hand to formation of $NO_2$ on the other hand to ozone depletion.

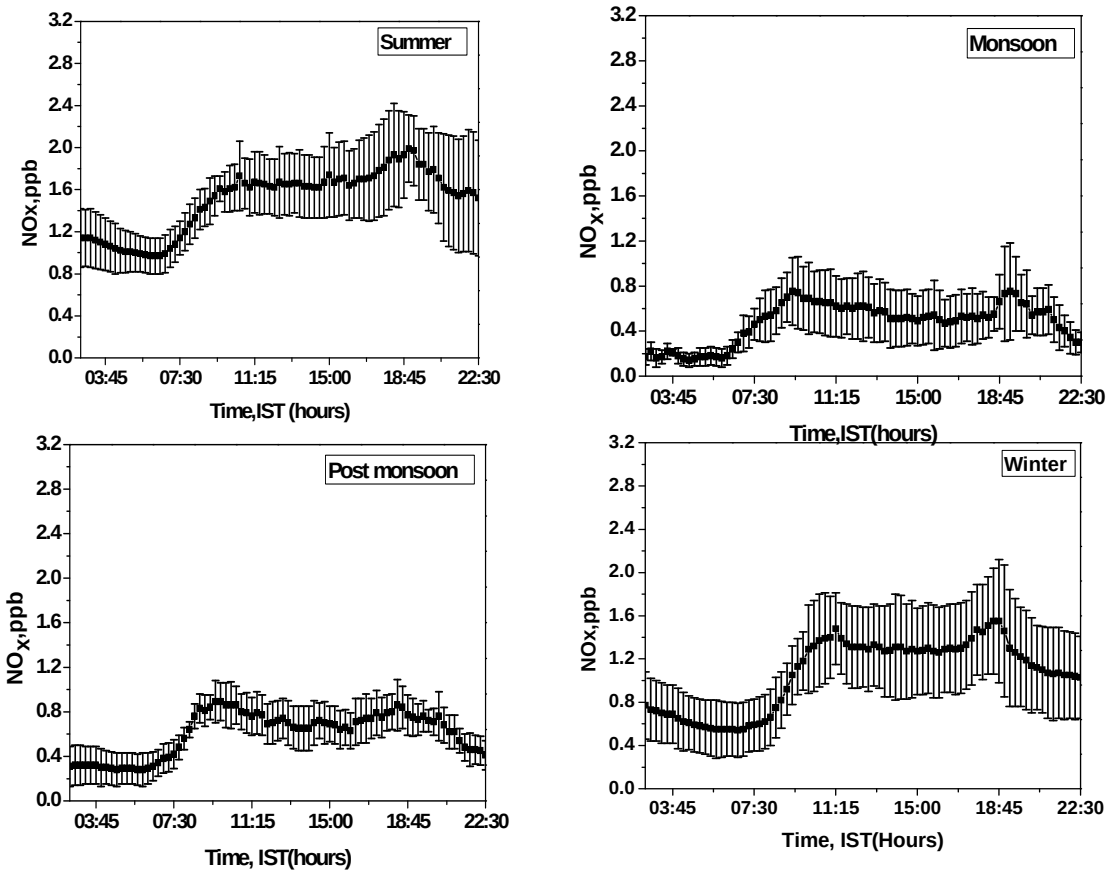


**Figure 8. Diurnal variation of NOx concentration during March 2010 to March 2012**
*(Vertical bars through the measurement points are standard deviation of the mean)*

The annual and seasonal variation of NOx concentration (Fig.9) was recorded during summer (1.60 ppb) and minimum in monsoon period (0.35 ppb). The monthly maximum during summer months such as March and April recorded 1.59 and 1.85 ppb, respectively and the minimum value of about 0.19 ppb observed in the month of August. The maximum concentration of NOx in winter and summer is mainly due to the anthropogenic activities around the observational site.

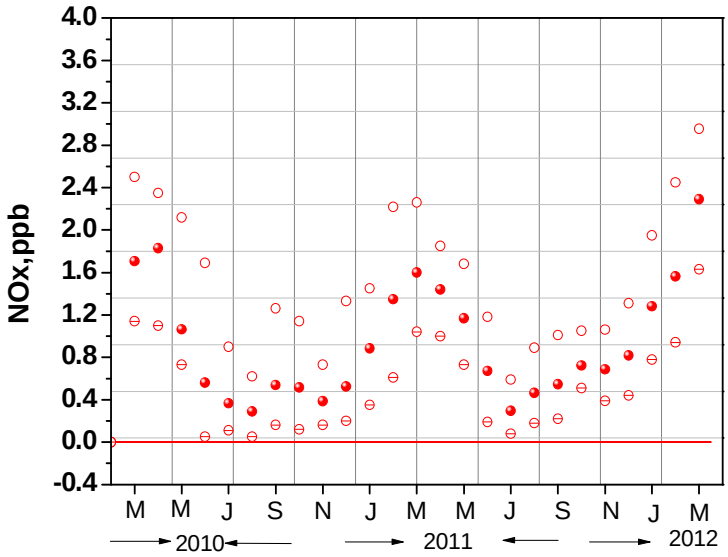


**Figure 9. Temporal variation of NOx**

***Comparison with other higher altitude stations***

Ozone concentrations measured in two different high altitude locations in India and the monthly mean minimum and maximum levels of surface ozone measured at different locations in India are listed in Table 1& 2. The comparison is aimed at illustrating the seasonal variation in ozone at two other high altitude sites (Mt.Abu, 24.6°N, 72.7°E,1680 m amsl and Nainital, 29.37°N,79.45°E, 1958 m amsl, [23, 15]. The ozone mixing ration is comparable or even higher during March (summer) when compared with Mt.Abu and Nainital, but the ozone mixing ratio is lower in June, July and August is highly comparable and its gradually increasing during winter is noticeable at three higher altitude sites.

**Table:1 Monthly variation of Ozone concentration measured at two different high altitude station in India.**

| Months | Mt.Abu | Nainital | Ooty |
|---|---|---|---|
| January | 46.2 | 37.3 | 43.38 |
| February | 46.7 | 43.8 | 43.82 |
| March | 42.6 | 56.6 | 61.87 |
| April | 38.7 | 63.1 | 51.04 |
| May | 37.9 | 67.2 | 44.27 |
| June | 25.4 | 44.0 | 22.30 |
| July | - | 30.3 | 20.80 |
| August | 25.1 | 24.9 | 17.10 |
| September | 37.1 | 32.0 | 20.19 |
| October | 46.8 | 42.4 | 30.50 |
| November | 48.8 | 43.9 | 30.35 |
| December | 44.4 | 41.6 | 36.60 |

***Correlation between ozone with temperature, relative humidity, rainfall and net incoming solar radiation***

The formation of ground level ozone depends on the intensity of solar radiation, the absolute concentrations of NOx and VOCs [24]. A large number of observations have shown that the concentrations of ozone increases with increase in the intensity of radiation and temperature on the clear days. The meteorological parameter such as temperature, humidity, rainfall and net incoming solar radiation for the period of March was high with low humidity level which shows a positive and negative correlation with ozone concentration. The highest ozone concentrations of the year generally occur during summer at this site, when sunlight is most intense. On a daily cycle motor vehicle activity rises throughout the morning, concentrations of NOx also rise due to the tourism and the high temperature and low humidity observed during dry summer months are favourable for ozone formation, whereas the condition is reverse during monsoon. The high relative humidity, low temperature and incoming solar radiation might attribute to low ozone concentration during monsoon seasons [39]. Nair *et al.*, [21] revealed that surface air temperature which is a measure of solar insulation, leading to higher photochemical reaction. In monsoon months, comparatively low value of $O_3$ is attributed to low UV radiation intensities and low ambient temperatures [1]. On the other hand, relative humidity which was higher in the rainy season shows negative correlation with temperature and ozone mixing ratio

and ozone level variation cycle depends on intensity of sun's radiation and on the temporal variation emission level of ozone precursors [18].

**Table: 2 Comparison of monthly mean minimum and maximum levels of surface ozone (O3) and NOx measured at different locations in India**

| Location | Site description | Altitude (m) | $O_3$ (ppbv) | NOx (ppbv) | Reference |
|---|---|---|---|---|---|
| Ahmedabad (23°N, 72.6°E) | Urban and Industrial | 49 | 16.8–51.4 | 4-19 | Lal *et al.*, 2000[16] |
| Delhi (28.7°N,77.2°E) | Urban and industrial | 220 | 5–20 | --- | Jain *et al.*, 2005 [14] |
| Pune (18.5°N, 73.8°E) | Urban | 559 | 12–54 | --- | Beig *et al.*, 2007[3] |
| Kolkata (23.4°N, 88.2°E) | Urban and industrial | 5 | 5–65 | 20-110 | Purkait *et al.*, 2009[30] |
| Dayalbagh (27.1°N, 78°E) | Urban | 169 | 10–75 | 15-40 | Singla *et al.*, 2011[38] |
| Anantapur (14.6°N, 77.6°E) | Semi-arid | 331 | 28.5–56.1 | 3.2-13.1 | Reddy *et al*,2011[40] |
| Gadanki (13.5°N,79.2°E) | Rural | 375 | 18–34 | 1.7-3.0 | Naja and Lal, 2002[22] |
| Joharapur (19.3°N, 75.2°E) | Rural | 474 | 15–44 | --- | Debaje *et al.*, 2006[8] |
| Trivandrum (8.5°N, 77.0°E) | Costal | 61 | 11.5–28 | --- | David and Nair, 2011[6] |
| Mt. Abu (24.6°N, 72.7°E) | High altitude | 1220 | 25–49 | 0.5-3.0 | Naja *et al.*, 2003[23] |
| Nainital (29.3°N, 79.45°E) | High altitude | 1958 | 25–67 | --- | Kumar *et al.*, 2010[15] |
| Ooty (11.2°N, 76.43°E) | High altitude | 2550 | 17-61 | 0.2-2.5 | Present Study |

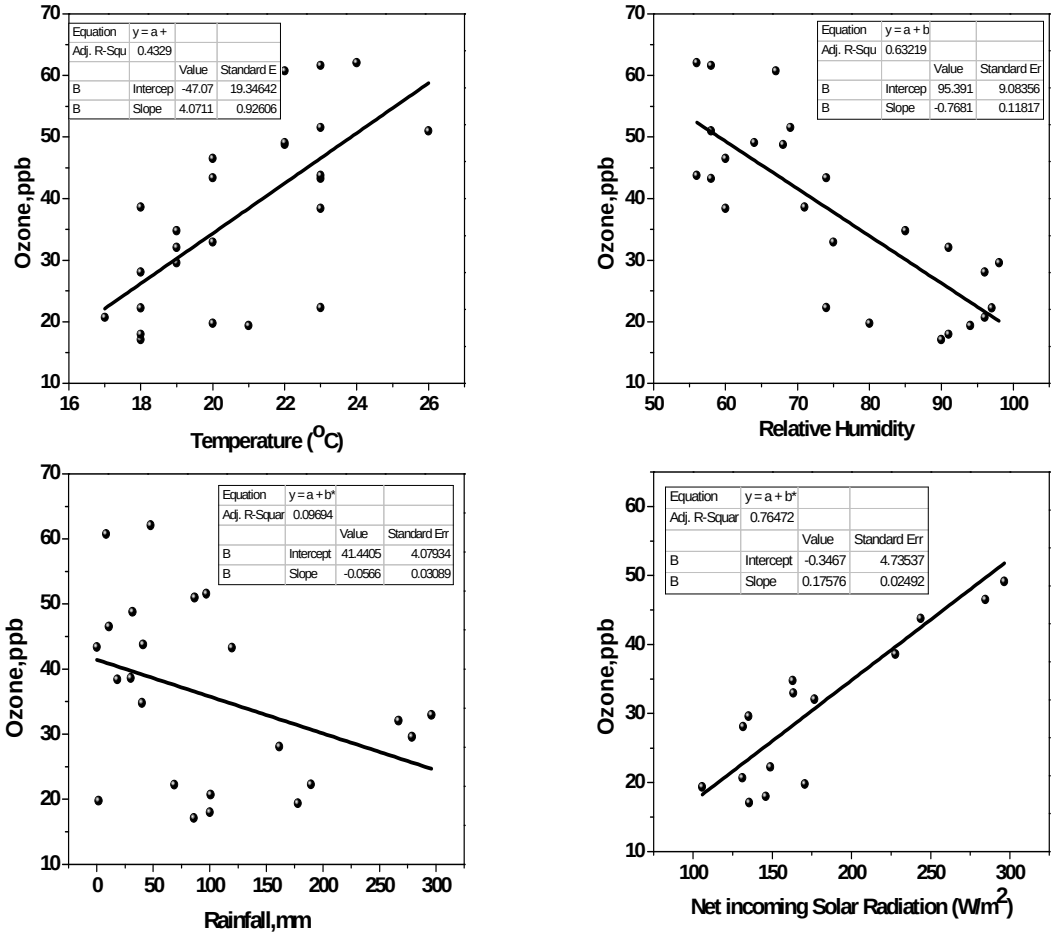


**Figure10. Correlation of Ozone with Temperature, Relative humidity, Rainfall and Solar flux**

The correlation between mean monthly temperature and monthly ozone was 0.42 (Fig.10). Net incoming solar radiation and mean monthly ozone showed a positive correlation ($r^2$ = 0.76). This indicates temperature and solar heating dependent ozone formation resulting in high ozone maxima during summer and winter season as compared to the monsoon and post monsoon season. The high solar flux and low humidity observed during the summer month are favourable for ozone formation. Relative humidity and mean monthly ozone concentration showed a negative correlation ($r^2$ = -0.63) and the trend was also followed for rainfall which showed negative correlation with mean monthly ozone ($r^2$ = -0.09). Whenever there is an increase in the relative humidity, the ozone concentration decrease, the negative correlation is attributed to the role of humidity in increasing the chemical interaction for the gases and photochemical smog [35].

Ozone production in nonurban areas is primarily controlled by nitrogen oxide (NOx) concentrations. A strong relation between ozone and oxides of nitrogen exhibited at this site ($r^2$ = 0.84). Whenever, the ozone concentration increases simultaneously the oxides of nitrogen starts decreasing and vice-versa. Nitrogen oxides play a predominant role in photochemical production of ozone in the troposphere [17]. Using this relation, estimated maximum ozone mixing ratio corresponds to NOx which illustrates the ozone destruction by titration of NOx is observed which results in lower mixing ratio during daytime but it is higher at night. A negative diurnal variation observed between these two pollutants is identified throughout study period.

## CONCLUSION

Using Ozone and NOx concentrations for a period of 2 years (March 2010 to March 2012), over the southern part of India has been characterized. The seasonal change in the NOx meteorological parameters, boundary layer processes, advection pathways and distinct source regions mainly dominate the observed seasonality in ozone characteristics over Ooty. The major findings are:

- The diurnal pattern of ozone was influence by the absence of day time buildup throughout the year. The pattern characterized by high ozone level during night time while, low level during day time.
- The seasonal mean values (ppb) of ozone for the 2 years period are 53.45 ± 8.18, 19.81 ± 2.4, 27.2 ± 5.67 and 42.81 ± 5.65 respectively for summer, Monsoon, post monsoon and winter.
- A monthly maximum NOx value was observed during summer month of April (1.85 ppb), whereas minimum value was observed during monsoon month of August (0.19 ppb).
- Low rainfall, higher temperature and solar radiation have given rise to the gradual build up of ozone during summer and winter. The unique meteorology over this region seems to play an important role in seasonal as well as diurnal variation of ozone.
- The strong diurnal and seasonal mean values of ozone during summer is attributed mainly to the ABL dynamics, advection pathways and source types.
- HYSPLIT back-trajectory analysis indicates the dominant easterly advection contributing to the seasonal high value of ozone during summer season

**Acknowledgement**
This work is supported by the Indian Space Research Organization (ISRO), Bangalore through its Geosphere Biosphere Programme (GBP) under AT - CTM project. The authors are grateful to Programme Director, ISRO – GBP, Bangalore for his help and constant encouragement.

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