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Diurnal and Seasonal Variations of Surface Ozone with Meteorological Parameters at Urban Site, Balabackya Nagar, Tirunelveli, Tamil Nadu, India

B.P.Usha[1], T. Chithambarathanu[2], R. Krishna Sharma[3]

[1] Associate Professor, PG and Research Department of Physics, Sri Paramakalyani College, Alwarkurichi, India, [2] Principal, S.T. Hindu College, Nagercoil, India,
Assistant Professor, PG and Research Department of Physics S.T. Hindu College, Nagercoil, India

Abstract- **Surface Ozone (Surface O3) measurement at the southern urban site of India, Balabackya Nagar (8.7314° N, 77.7081° E), Tirunelveli, Tamil Nadu was carried out for the first time for a period of one year from November 2016 to October 2017. The increasing trend of surface O3 is a threatening effect to be monitored seriously as the effect worsen the life of biotic and abiotic factors on the earth. The surface O3 concentration for various seasons were measured using a portable gas sensitive semiconductor sensor based monitor. From the measurements it was observed that the ozone concentration increases from morning to noon and then decreases. The peak value was obtained in the afternoon. The variation of surface O3 with meteorological parameters like temperature, Relative Humidity, wind speed were also studied. From the measurements it was revealed that the temperature shows a positive correlation, Relative Humidity shows a negative correlation whereas wind speed shows positive correlation except during winter season with surface O3. In the observation site the surface O3 variation was recorded seasonally. A diurnal variation pattern was obtained for surface O3 in the site. Maximum value of surface O3 recorded was 51ppb during summer and the minimum value of 6.5 ppb during winter season. The seasonal averaged value of surface O3 was 49.72ppb during summer and a minimum of 10.42ppb during winter. The maximum rate of change of ozone was obtained as 4.67ppb.**

Keywords: Surface O_3, meteorological parameter, correlation, diurnal variation.

INTRODUCTION

Ozone an allotrope which has three oxygen atoms bound together and forms the chemical formula O_3 is present in stratosphere producing positive effects but harmful when present in the troposphere. The expansion of industrialization and increasing population resulted the huge demand of fossil and bio fuels combustion, prominent energy demand and higher agricultural activities induce enormous emission of pollutants into the atmosphere [1]. According to the intergovernmental panel on climatic change (IPCC) 2001, the surface O_3 is considered as the third most powerful greenhouse gas in the atmosphere after CO_2 &CH_4 with radioactive forcing of $+0.35 wm^{-2}$. Each addition of O_3 mole in the atmosphere is 1200-2000 times more effective than global warming [2]. The ozone related deaths during June to August of 2000, 2001 and 2002 in Netherlands were 990, 1140 and 1400 respectively [3]. Tropospheric ozone has two main sources namely stratosphere intrusion affecting surface ozone [4] and in-situ production via photochemical oxidation of carbon like compound (CO, CH_4 & VOCs) in presence of NOx. Tropospheric ozone has a detrimental effect on human health and ecosystem productivity [5]. The life time of ozone varies from season to season with altitudes between a few days at the surface to a few weeks in the troposphere. The main sources for surface O_3 enhancement are photochemical production, atmospheric transport, chemical destruction, dry deposition on vegetation water and other materials and stratospheric-tropospheric exchange. It is evident that the percentage of stratospheric ozone is decreasing while that of tropospheric ozone is increasing. This increase in rate is 0.32% per year.[6]

The life time of NO_x varies with the photo chemical environment and is of the order of an hour to a day. Trace gas observations caught the attention of scientists in India in the beginning of 1990's Indian space Research Organization - Geosphere Biosphere program (ISRO - GBP) took an initiative to increase the awareness and the understanding of atmospheric trace gases, their chemistry, transport, pathways over the Indian sub continent. As a part of ISRO-GBP project, the continuous measurement of trace gases (SO2, NO_x, CO & O_3) were studied at the IIT, Kanpur. The variation of the trace gases and the relationship among them along with the role of meteorological parameters were reported [7]. Analysis of diurnal and seasonal behaviour of surface O_3 and its precursor

(NO_x) at a semi-Arid site in southern India was reported by Reddy et al at Ananthapur [8]. They have showed that the O_3 concentration was positively correlated with temperature and negatively correlated with relative humidity. On the contrast NO_x showed a negative correlation with temperature and positive correlation with relative humidity. In the selected site Tirunelveli, Southern most part of India nearby Kanyakumari District the surface ozone measurements were carried out for the first time. The seasonal variation of surface O_3 with meteorological parameters were observed along with the variation of the main precursor of ozone, NO_2.

2. SITE DESCRIPTION WITH GENERAL METEOROLOGY

Pilot study was carried out in and around Tirunelveli and accordingly Balabakya Nagar was chosen as the study site. The selected site Balabakya Nagar (8.7314°N, 77.7081°E) is situated to the north of Tirunelveli Junction bus stand and east of junction railway station which is a heavy traffic prone area. Tirunelveli is the administrative head quarters of Tirunelveli District. It is located at the southern most tip of Deccan Plataue. The city covers an area of 108.65km^2. It has an extensive transport network. The boundary of the study area is Thatchanallur at the north Tirunelveli Junction bus stand at the south, Tirunelveli Junction Railway Station at the west and Madurai Bypass road at the east.

The climate of Tirunelveli is hot and humid. The average temperature during summer ranges from (25°C - 41°C) and during rest of the year it is (18°C - 29°C). The average annual rain fall is 680mm (weatherundergrounds.com). The month of May is generally hottest in the interior parts of the district. In the month of May and June weather remains hot and the maximum temperature reaches upto 45°C.

As per the climatic description of Tirunelveli District the seasons of the district are Winter, Summer and Monsoon.

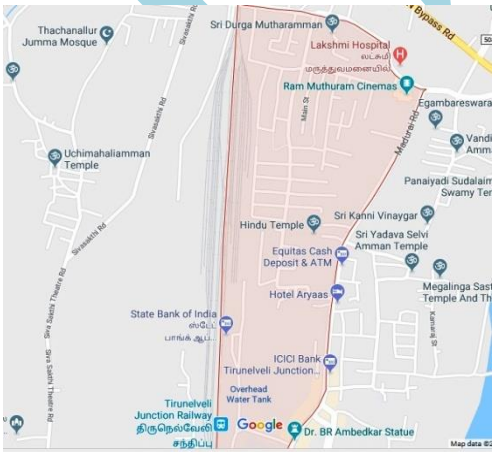

Fig-1 location of the measurement site

3. AIM OF THE STUDY

Global warming and environmental pollution are threatening problems to be seriously discussed. Pollution causes damages to biotic and abiotic factors of the biosphere. This study is being carried out with special attention focused on the diurnal and seasonal variations of surface O_3 over an urban site of Tirunelveli on the changing climate. Apart from the natural phenomena the manmade activities pollute the environment specifically the troposphere with threatening effect of surface O_3. Variation of O_3 with meteorological parameters (Relative humidity, Wind speed and temperature) is also discussed in the study.

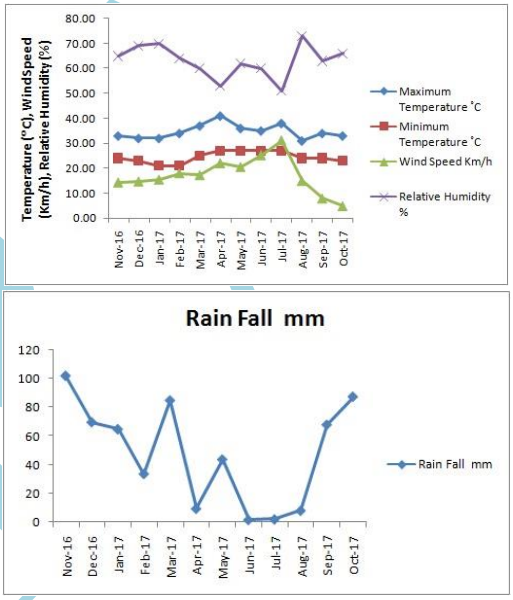

Fig-2 Monthly average values of Meteorological parameters

Fig.2, Shows the monthly average variations of relative humidity, wind speed, maximum temperature, minimum temperature and rainfall. The minimum average temperature recorded is 21°C (July & December) and the maximum average temperature recorded is 41°C (April) During the observation period in the selected site, it was observed that the Relative humidity was highest in the month of August (75%) and lowest in July (51%). Wind speed is maximum in August (20.52km/h) and minimum in November (2.99km/h) from Fig: 2 it is noted that the site is influenced by the monsoon rainfall (October and November) according to the information from weatherundergrounds.com

3. METHODS

Surface ozone variation for a period of one year from November 2016 to October – 2017 were carried out in the site by using a portable gas monitor aeroqual S300 coupled with ozone and NO_2 sensor. The observation period covers three seasons, winter - 2016 – 17, summer 2017, monsoon 2017.

Fig 3 : Aeroqual Ozone Monitor S300

The instrument is portable, reliable and comparatively cheap and the gas concentration can be obtained directly. It is calibrated against a certified UV photometer. This instrument was used by many researchers for measuring atmospheric ozone. The observations were taken for all possible days of the months starting from 5:30 hours morning to 5:30 hours next day morning with an interval of 3 hours comprising 9 readings. Fig 3 shows the Aeroqual S300 monitor with ozone and NO_2 sensor heads.

4. RESULTS AND DISCUSSIONS

The observed ozone variations were analyzed diurnally and seasonally. Daily average was analyzed from hourly basis and monthly average is analyzed from daily values seasonal average was analyzed from monthly values.

4.1 Frequency distribution

Frequency distribution is a representation which displays the number of observations within a given interval.

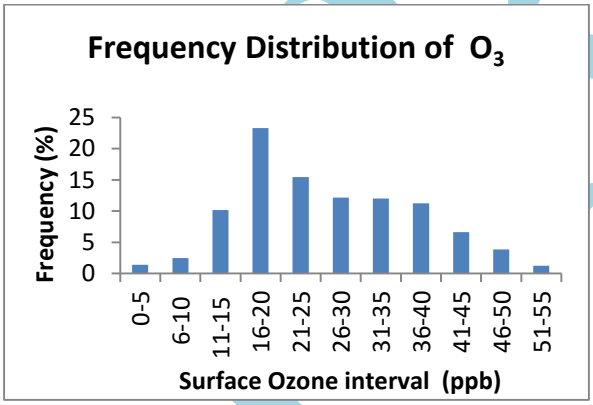

Fig.4 : Frequency Distribution of Surface Ozone

Fig:4 shows the frequency distribution of surface O3 for different concentration ranges.

From the observed data points, nearly 62% of the data lies between (0-30) ppb. The highest distribution is between (16-20) ppb. The frequency distribution of surface ozone recorded by previous researchers in Nagercoil site showed that 70% data lies between (5-25) ppb [9]. In Kanyakumari it was recorded that 60% of data between (11-30) ppb [10]. From Fig.4, the frequency distribution is as follows,

i) 4% in (0-10) ppb
ii) 61% in (11-30) ppb and
iii) 35% in (31-55) ppb

The maximum distribution is 23% in (16-20) ppb and the minimum distribution is (1.23) % in (51-55) ppb

4.2 Diurnal variation of surface ozone concentration

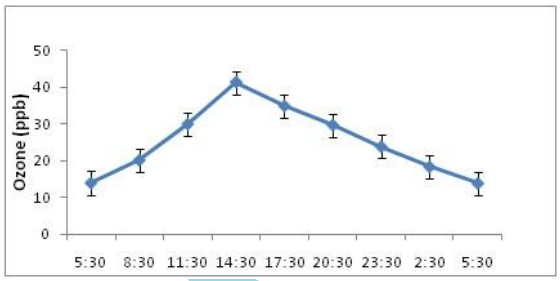

Fig.5 : Diurnal Variation of Surface O3 during the period Nov 2016 – Oct 2017

In general diurnal variation stands for the ozone fluctuations that occur during each day. It is very useful in understanding the various processes that are responsible for the production and loss rates. The diurnal variation is a measure of overall budget of production and loss rate. The diurnal variation is characterized by a broad minimum at night, a rapid rise in the morning after sunrise and sharp maximum near noon.

The overall averaged diurnal variation for the period of one year (November 2016 – October 2017) is shown in Fig 5. The variation is directly related to the solar intensity through photochemical reactions. The maximum ozone concentration is observed mostly around 14:30 hours in the afternoon. The minimum value is observed in 5:30 hours in the morning. This is because accumulation of precursor gases in the morning hours and late evening hours.[11] The low concentration of surface O_3 observed in the morning hours is due to the lower boundary layer height which mainly reduces the mixing process between ozone poor surface layer and ozone rich upper layer. The mixing ratios of ozone start increasing gradually after sunrise, attaining maximum values during local noontime.[12] Apart from the role of photochemistry, boundary layer meteorology and dynamics also play a key role in ozone variability.[13] Boundary layer attains the maximum height during afternoon hours due to the increase in surface heating.

During this time, trace species gets vigorously mixed within, thus forming convective mixing layer. During the one year study period, a minimum of 6.5ppb at winter season and maximum of 51ppb at summer season were observed.

The increase of surface O3 concentration during daytime is attributed to the photolysis reactions of NO_2 and photo oxidation of VOC's CO, hydrocarbon and other ozone precursors. The downward transport of surface O_3 is by vertical mixing due to convective heating [14]. The ozone concentration is reduced as the night inversion layer is formed in the evening and no markable changes occur. At night there wouldn't be any radiation flux, no photolysis and hence no photochemical reaction takes place and thereby O_3

concentration is reduced. Also, the production rate of O_3 in the morning is higher when compared to the destruction rate of O_3 in the evening.

In the process of NO_x formation one O_3 is removed per emitted NO. But in O_3 formation, four or more O_3 is formed per NO emission [15]. The diurnal variation of O_3 at this site could be explained with the atmospheric process associated with the meteorological parameters.

4.3 Seasonally averaged diurnal variations of O_3

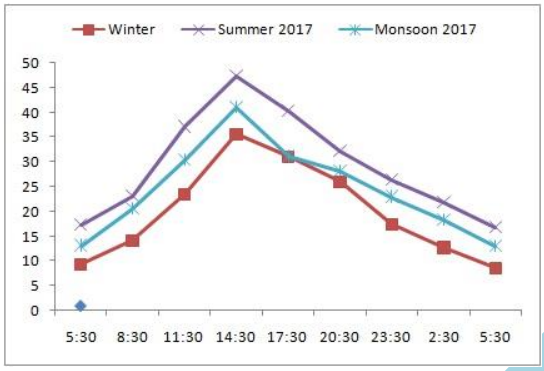

Fig.6 Seasonally averaged diurnal variations of O_3

The diurnal variation of average ozone concentration for various seasons (Summer 2017, Monsoon 2017, Winter 2016-17) are depicted in fig.6 surface O3 concentration is in peak in the summer with an average of 47.34 ppb. It is clearly due to the direct incoming solar radiation. The high temperature has direct influence on chemical kinetic rates and the mechanism leads the O_3 production[16]. Previous studies reported O3 temperature relationship disclosed O_3 formation from Preoxy Actyl Nitrate (PAN) decomposition as temperature increases ~ 37°C.

The maximum and minimum values of O3 in various seasons obtained in the observation site is as follows,

Table. 1 : Seasonal Maximum and Minimum Ozone Concentration

Season	Maximum O_3 (ppb)	Minimum O_3 (ppb)
Winter 2016 – 17	35.58	9.46
Summer 2017	47.34	17.32
Monsoon 2017	39.68	12.99

The overall maximum O_3 concentration was observed in the summer and the minimum during winter. The factors like the meteorological parameters temperature, wind speed, wind direction, relative humidity and the formation of precursors like NO_x, CO. CH_4, VOC's and hydro carbon are responsible for the variation in the surface O3 concentration. The reason for the minimum surface O3 concentration obtained in the winter is the reflection of radiation back to the stratosphere by the cloudy skies and the rain which gets increased in this season makes the precursors reduced. As the temperature decreases in the winter, the thermal decomposition of O_3 precursors is reduced. The O_3 concentrations obtained were comparable to those reported at other urban location in India [17]. But it is very low when compared to the rural and high altitude areas.

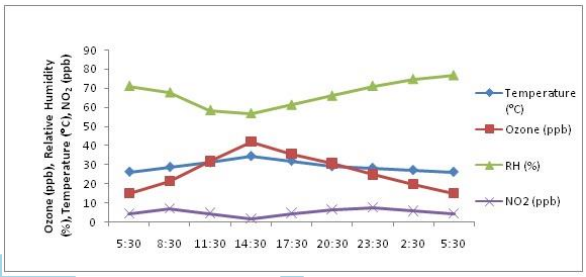

Fig. 7 : Variation of Ozone with Temperature, Relative Humidity and NO_2

NO_x is the main precursor of O_3 and it shows the variation as in the above figure. Fig.7 shows that when O_3 reaches a peak in the afternoon, the NO_2 shows the lowest concentration. This proves the O_3 production via photochemical oxidation of carbon like compounds (CO, CH_4, HC's) by reacting with OH radicals in presence of NO_x [18].From Table-1, it is noted that the maximum and minimum values of surface O_3 are highest in the summer. Likewise the minimum values of surface O_3 in winter (Both the maximum and minimum values of surface O_3) From fig7, it is very clear that surface O3 and temperature variations are direct and surface O_3 and temperature variation with RH and NO_2 are indirect.

Table-2 : Comparison of trace measurements

Location	Lat (deg.)	Lon. 9deg.)	Site Type	Period	Concentration of		Reference
					NO_2 (ppb)	O_3(ppb)	
Tirunelveli	8.71	77.76	Urban	11/2016-10/2017	5.21 ± 2.2	26.16 ± 2.07	This Study
Agra	27.18	78.02	Urban	01/2002-12/2002	-	21.0	(Satsangi et al-2004)
Ahmedabad	23.03	72.58	Urban	06/2003-05/2004	9.5 ± 2.7	20.7 ± 5.5	(Beig et al-2007)
Ananthapur	14.68	77.60	Rural	01/2010-12/2010	5.1 ± 0.7	40.7 ± 3.7	(Reddy et al-2013)
Kannur	12.87	74.90	Coastal Rural	11/2009-10/2010	2.5	18.4 - 44	(Nishanth et al-2012)
Kanyakumari	8.4	77.32	Coastal	03/2012-02/2013	4.77	28.81	(Krishna Sharmal & Chithambara Thanu-2013)
Nagercoil	8.11	77.4	Urban	03/2009-02/2010	4.75 ± 0.68	20.8 ± 2.57	(Elampari & Chithampara Thanu 2010)
Pune	18.53	73.86	Urban	06/2003-05/2004	-	30.9 ± 14	(Beig et al-2007)
Thumba	8.52	76.87	Coastal Rural	04/1997-03/1998	-	23.0	Nair et al- 2002)

The table-2 depicted the results of measurements of trace gases (O_3 & NO_2) over many places of Indian subcontinent. The result of this study is comparable with the results of nearby district Kanyakumari. Even though the two districts are nearby there exists a vast difference in the climate.

Tirunelveli - O_3 : 26.16 ± 2.07 ppb and NO_2 : 5.21 ± 2.2ppb.
Nagercoil - O_3 : 20.08 ± 2.57 ppb and NO_2 : 4.75 ± 0.68 ppb.
Kanyakumari - O3 : 28.81 ppb and NO_2 : 4.77 ppb.

From table 3, it is clear that the study site (Tirunelveli) 77.76° longitude and Ananthapur 77.70° longitude has a concentration of 5.21 ± 2.2 ppb and 5.11 ± 0.7 ppb NO_2. Kanyakumari 77.32° longitude and Nagercoil 77.4° longitude has a concentration of NO_2 4.77ppb and 4.75 ± .68ppb. In the sites Tirunelveli, Agra, Kanyakumari, Nagercoil, Thumba the recorder values of surface O_3 concentrations are comparable in magnitude.

4.4 Rate of change of O_3 concentration for various seasons(dO3/dt)

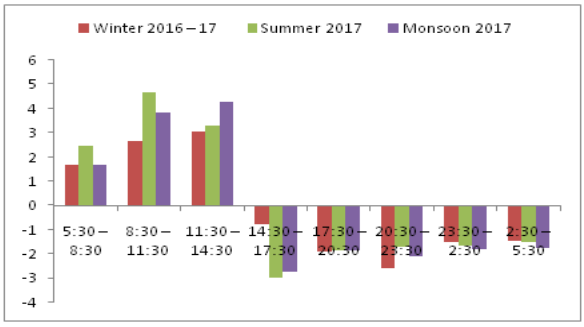

Fig. 8 (a) Rate of change of O_3 concentration for various seasons

Fig.8 (a) shows the rate of change of O_3 in the selected site. This parameter can be used as an indicator of urban and rural chemical environment [19]. Urban sites show a symmetric variation of O_3 in morning & evening rate of change whereas in rural sites asymmetric variation of O_3 is obtained. The O_3 build up rate is higher in morning than the loss rate in the evening.

Rate of Change of Ozone $\frac{do_3}{dt} = \frac{O_3(t_2) - O_3(t_1)}{3}$

Table.3 : Comparison of Rate of change of Ozone for various sites in India.

Location	Site Type	Rate of Change of O_3		Reference
		(08:00 – 11:00)h	17:00-19:00)h	
Tirunelveli	Urban	3.45	-3.11	This Study
Agra	Urban	2.5	-2.4	(Singla et al-2011)
Ahmedabad	Urban	5.9	-6.4	(Lal et al-2007)
Ananthapur	Rural	4.6	-2.5	(Reddy et al-2013)
Delhi	Urban	4.5	-5.3	(Ahammed et al. 2000)
Gadanki	Rural	4.6	-2.6	(Naja Lal.2002)
Kanpur	Urban	3.3	-2.6	(Abhishek Gaur et al,2014)
Kannur	Coastal Rural	4.9	-6.4	(Nishanth et al-2012)
Kanyakumari	Coastal	4.6	-3.9	(Krishna Sharmal et al, 2013)
Nagercoil	Urban	4.2	-2.4	(Elampari & Chithampara Thanu 2010)
Pune	Urban	4.8	-2.6	(Shendhe et al-2007)
Thumba	Coastal Rural	5.5	-1.4	Nair et al- 2002)

From the table 3 it is clear that the four rural sites showed an asymmetric variation of surface O_3 in the buildup and loss rate whereas all the urban sites showed symmetric variation. This selected area is an urban site which shows a symmetric variation of surface O_3 in buildup and loss rate with 3.45ppb /h and -3.11ppb/h.

From fig.8, it is clear that the highest seasonal rate of change of O_3 observed in the selected site is +4.67 ppb which is very close the recorded values of Nagercoil. +4.66 ppb and Kanyakumari +4.2 ppb [20]. Irrespective of the season, the night time rate of change of O_3 is negative which is due to O_3 loss to surface deposition.

4.4 Surface O_3 and meteorological parameter

The surface O_3 concentration variation is strongly associated with the meteorological parameters like, temperature, RH, WS, etc.

The O_3 concentration fluctuates with the metrological parameters. This variations and the correlation of these parameters with surface O3 in our site is discussed in the following sections.

4.4.1. Correlation between relative humidity and surface O_3

RH is used very often in air pollution studies. If the humidity is high, the photochemical reaction for the O_3 removal will be enhanced. The higher atmospheric instability slowed down the photochemical process and the surface O_3 is deposited on the water droplets and thereby depleted. It was observed that there exist a negative correlation between RH and surface O_3 in all seasons. The values of correlation co-efficient between relative humidity and surface O_3 for various seasons are displayed in Table. 4. The overall value of correlation co-efficient between RH and surface O3 in -0.5623.

Table.4 : Correlation between O_{3avg} and RH_{avg}

Season	Correlation Co-efficient	P Value
Winter – 2016 – 2017	- 0.4821	0.0056
Summer – 2017	- 0.6834	0.0000
Monsoon – 2017	- 0.5217	0.0027

4.4.2 Correlation between temperature and surface O_3

The O_3 concentration is maximum when the temperature is at peak in most of the days. The temperature varies within 24 hrs of a day and fluctuate at different seasons. The maximum observed temperature is 41°C (summer) and the minimum is 21°C (Winter) in the selected site. Irrespective of the seasons, the peak value of temperature recorded was at 14:30 hours. From Fig.7 shows the variation of surface O_3 and temperature. The correlation coefficient is the in all seasons. The values are shown in the Table 6.

Table – 5 : Correlation between O_{3avg} and T_{avg}

Season	Correlation Co-efficient	P Value
Winter – 2016 – 17	0.78	5×10^{-20}
Summer – 2017	0.69	3×10^{-16}
Monsoon – 2017	0.72	6×10^{-19}

The overall value of Correlation co-efficient between temperature and surface O_3 is 0.73.

4.4.3 Correlation between wind speed and surface O_3

The wind speed is an important factor which carry the O_3 transport from stratosphere and the transport of ozone precursor from industries etc., in the selected site during the observation period, the average wind speed varies from a minimum of 2.99 km/h to a maximum of 22km/h. The values of the correlation coefficient for various seasons are given in Table 7.

Table.6 : Correlation between W_{avg} and O_{3avg}

Season	Correlation Co-efficient	P Value
Winter – 2016 – 17	- 0.2449	0.0201
Summer – 2017	0.6321	0.0013
Monsoon – 2017	0.5238	0.0187

6. CONCLUSION

From the result of the study carried out at Tirunelveli, the southernmost tip of India nearby Kanyakumari, the variation of surface O_3 concentration for a period of one year the following inferences are made. This study is carried out in the site as a first time where no previous studies had been carried out.

❖ Continuous observation of surface ozone variation is carried out at the observation site, Tirunelveli for one year (3 Seasons). The observed results are,

❖ The diurnal variation of surface O3 concentration shows its reliance mainly on photochemical reactions.

❖ While analyzing the frequency distribution of data points in the site, i) 4% distribution in (0-10) ppb, ii) 61% in (11-30) ppb, iii) 35% in (31-55) ppb. The maximum distribution corresponds to (23.30) % in (6-20) ppb and the minimum distribution (1.23) corresponds to (51-55)ppb. From the observation it is clear that the environment experiences comparatively low concentration of surface O_3 for a longer period of time.

❖ The metrological parameters are responsible for the O3 precursors production and destruction processes during the study period and the seasonal variation of surface O3. The surface O3 concentration observed is maximum in summer followed by monsoon and minimum in winter.

❖ The correlation between surface O3 and RH is negative in all seasons and the correlation between surface O3 and temperature is positive is all seasons and the correlation between surface O3 and wind speed is the positive except in winter.

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