Indian Journal of Radio & Space Physics
Vol 45, June 2016, pp. 79-86

Variation of ozone with meteorology in surface air over two sites of southern Tamil Nadu, India

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Received 13 March 2015; revised 18 May 2016; accepted 23 May 2016

Ozone is an important atmospheric constituent due to its role both as a greenhouse gas and an oxidant. Ozone can be good or bad depending upon where it resides. Interactions between ozone and climate naturally occur not only in the stratosphere, but also at the earth's surface (troposphere). In this study, the amount of surface ozone (SOZ) concentration in air was measured at the two sites of southern Tamil Nadu, viz. Kottar (8.1739°N, 77.4389°E) and Suchindrum (8.1550°N, 77.4650°E)] influenced by almost same meteorological conditions for one year from February 2014 to January 2015 using a portable Aeroqual S200 and S300 monitor. A lucid global day-night variation was observed for SOZ concentration. Summer months recorded the maximum SOZ of 56 ppb. The least values of SOZ were observed during northeast monsoon for both the sites. The maximum rate of change of SOZ was found around 4.6 ppb, which can be compared well with Delhi and Gadanki. A difference of 2.01 ppb was observed in the mean of daily average surface ozone concentration between the two sites. Statistical t-test was performed in this study, which revealed that this difference is due to the difference in the levels of precursor gas emissions and vegetation at the two sites.

Keywords: Climate, Greenhouse gas, Surface ozone, Air quality

PACS Nos: 92.30.Np; 92.60.Sz; 92.70.Cp

1 Introduction

Tropospheric ozone, which is also known as ground level ozone (GLO) or surface ozone (SOZ), is very harmful and a less known secondary pollutant. Ozone (O_3) in the lower atmosphere, particularly surface ozone, is highly variable in space and time[1]. The changes in the concentrations of O_3 have increased the trapping of terrestrial radiation in the atmosphere and thereby, enhanced the greenhouse effect above its natural level[2]. It plays an important role in global warming because of its strong absorption band, centered at 9.6 µm (Ref. 3). It is an important air quality issue and causes serious health problems and damage to materials and ecosystems[4]. It is considered as an important pollutant by the Environmental Protection Agency (EPA) and World Health Organization (WHO). Ozone is a powerful oxidant and as such it can react with a wide range of cellular components and biological materials. Epidemiological and toxicological studies indicate that higher concentration of ozone in the boundary layer is harmful to biological health[5] and several types of ozone related short-term health effects have been detected so far. Ozone plays an important role in the energy budget of the atmosphere, since it absorbs both solar and infrared radiation[6]. Being a main atmospheric oxidizing component and an optically active constituent, it plays an important role in the climate system of the earth[7]. An increase in the tropospheric ozone causes increase in the surface temperature. The atmosphere contains nitrogen (N_2) and oxygen (O_2) as major constituents. However, there are many other gases, which are in low quantities called trace gases. Trace gases play a great role in atmospheric composition, chemical reaction, radiative interaction, climate change and pollution, even if their abundance is very low. Therefore, knowledge of these trace gases is important to understand and quantify the atmospheric changes, such as global warming, atmospheric pollution and change in the chemical composition of the troposphere.

Ozone is one of the important trace gases in the photochemistry of the atmosphere and it can be considered as an indicator of the overall burden of the atmospheric oxidants[8]. The results of the study

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conducted by Deb Roy *et al.*[9] indicate the impact of ozone on crop yield in India. It is evident that the amount of stratospheric ozone is decreasing while the amount of tropospheric ozone is increasing[10]. Atmospheric CO_2 concentrations are currently rising at a rate of approximately 0.5% per year and surface ozone (ground level O_3) values are increasing at a rate of 0.32% per year[11]. Ozone concentration in the city of Ahmedabad is increased by 1-2% per year[12]. In Pune city, during 2001-2005, concentration of ozone was reported to be as high as 180 μg m^{-3} and the permissible limits were repeatedly breached[13]. Many researchers have reported an increasing trend in surface ozone concentrations in India[14-16].

2 Photochemistry of Surface Ozone

The majority of tropospheric ozone formation occurs when nitrogen oxides (NOx), carbon monoxide (CO) and volatile organic compounds (VOCs) react in the atmosphere in the presence of sunlight. NOx, CO, and VOCs are called ozone precursors.

$$NO_2 + h\ (\ <= 430nm) \rightarrow NO + O \quad \ldots (1)$$

$$O + O_2 \rightarrow O_3 \quad \ldots (2)$$

But NO reacts relatively rapidly with ozone to form NO_2 under atmospheric conditions:

$$NO + O_3 \rightarrow NO_2 + O_2 \quad \ldots (3)$$

where, the photolysis rate of NO_2 is J1 and the rate of reaction (3) is k. In one-dimensional steady state condition, ozone concentration is given by:

$$[O3] = J1[NO2] / (k\,[NO]) \quad \ldots (4)$$

If each side of equation is divided by $[O_3]$, the right hand side is termed as the Leighton ratio (Φ) (Ref 17):

$$\Phi = j_{NO2}[NO_2] / (k_1[\,NO]\,[O_3]) \quad \ldots (5)$$

Reactions (1), (2) and (3) constitute a cycle with an overall net chemistry, which gives a steady state concentration of ozone.

3 The Present Study

The present study aims to assess the levels of SOZ over two sites of southern Tamil Nadu. The objective is to: (i) execute statistical analysis of ozone and meteorological parameters for a period of one year; (ii) analyze ozone concentration in terms of diurnal and seasonal variations; (iii) compare the variation of ozone between two selected sites influenced by similar meteorological conditions and find the difference in the mean; (iv) determine if there is any correlation; and (v) identify the most important factors in determining ozone levels.

3.1 Study area

The ozone concentration data, reported in this study, represent measurements taken at two types of sampling sites in Kanyakumari district. It was decided to choose one site, which is surrounded by healthy vegetation and free from vehicular emissions (Suchindrum S2) and another site near a major traffic thoroughfare (Kottar S1). The site S1 is dominated by vehicular emissions and commercial activities as compared to S2. Measurements were carried out for a period of one year from February 2014 to January 2015. Figure 1 shows the study area. The climate of the district is divided into four seasons (as per NIC and IMD). The study area witnesses its summer season from March to May followed by southwest monsoon that extends from June to September. Northeast monsoon is from October to December followed by winter season during January and February (http://tnenvis.nic.in/files/KANYAKUMARI%20%20.pdf).

3.2 Method

Surface ozone measurements were carried out using a portable sensitive gaseous monitor Aeroqual S200 and S300 coupled with ozone sensor. Figure 2 shows Aeroqual S200 monitor. Its ultra-low concentration ozone sensor head measures the ozone concentration from 0 to 150 ppb with an accuracy of ±1 ppb (from 0 to 100 ppb). The measurement unit being either ppb or μg m^{-3}. The operating temperature varied from -5°C to 50°C, relative humidity limits were 5% and 95%. The ozone sensor was calibrated against a certified UV photometer. Eight readings were taken per day starting from 0530 hrs LT to 0230 hrs LT. Aeroqual ozone monitors have been used for the measurement of ozone and nitrogen dioxide[11,13,18-22]. Recently, the Aeroqual Series ozone monitor was used to calculate ozone deposition on snow and ice surfaces at Scott Base in Antarctica by a research team from Auckland University in New Zealand (http://www.aozp.co.uk). O_3 concentrations were acquired by portable analyzers (Aeroqual Series 500), which make use of a semi-conductive sensor specific for detecting low O_3 levels (0–0.5 ppm)[23].

4 Results and Discussion

4.1 Diurnal variaton

The diurnal variation is characterized by a broad minimum at night, a rapid rise in the morning after

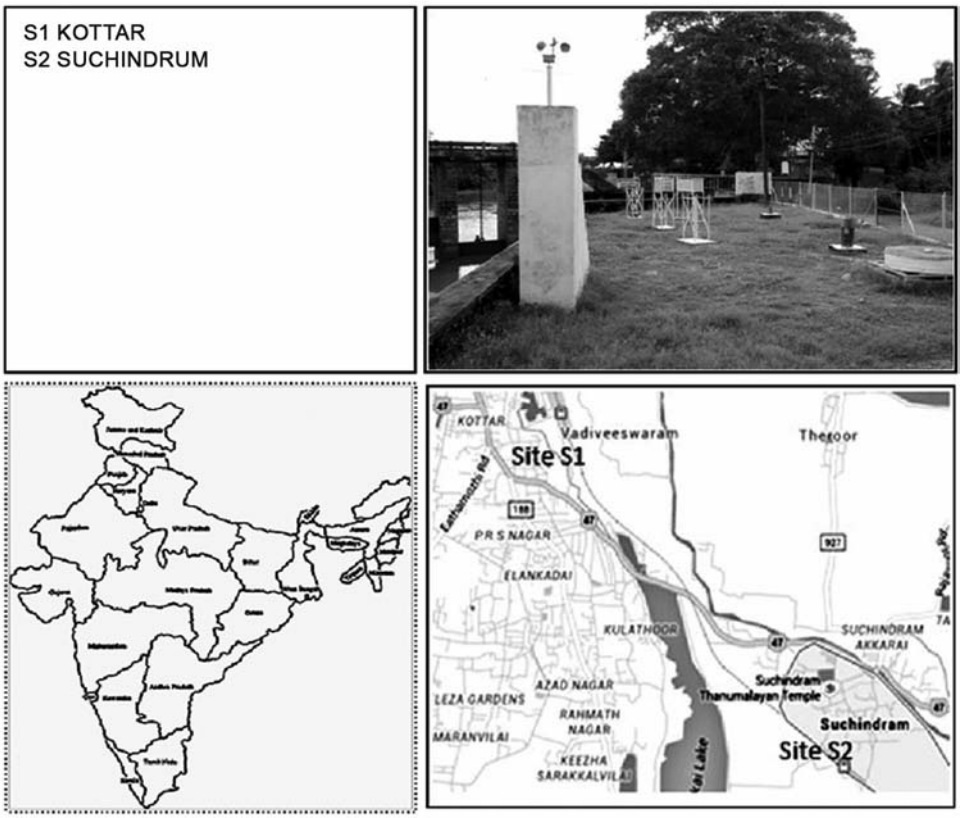

Fig. 1 — Sampling sites

Fig. 2 — Aeroqual S200 monitor with ozone sensor

sunrise and a sharp maximum near noon[24]. Surface ozone variations showed a clear diurnal pattern for both the sites. The ozone concentration gradually increased after sunrise because of the fact that photochemical reactions are dominant only in the presence of sunlight. The maximum value of surface ozone was recorded around 1430 hrs LT and this is due to the high intensity of solar flux during afternoon hours. This indicates that the photochemical production of surface ozone is strongly related to the surface temperature. During the late evening hours, a decreasing trend was observed in the surface ozone levels because the lower regions of the boundary layer become thermally stratified and stable as the surface cools; hence, greatly reducing entrainment of ozone from the free troposphere. Ozone levels were observed to be lower during nighttime when there is no O_3 production from photo oxidation of precursors. In addition, the titration of O_3 by NO in the shallow boundary layer and loss due to surface deposition continues during the nighttime. Also, this low concentration of ozone during night and early morning time can be a result of its deposition and surface chemical reactions[25]. The mixing ratios of ozone start increasing gradually after sunrise, attaining maximum values during local noon time[26]. Day time higher ozone levels are mainly due to the photochemical production of ozone[12]. Apart from the role of photochemistry, boundary layer meteorology and dynamics also play a key role in ozone variability[27]. Boundary layer attains the maximum

height during afternoon hours due to the increase in surface heating. During this time, trace species get vigorously mixed within, thus, forming convective mixed layer[28]. The lowest values in the morning (0530 hrs LT) are the results of night time ozone destruction process including loss due to atmospheric chemistry and dry deposition on the earth. But the steep increase observed in the forenoon hours is due to the domination of photochemical production of ozone in highly conducive weather. The overall diurnal variation of SOZ at both the sites is represented in Fig. 3.

4.2 Seasonal analysis

The seasonal variation is important for the better understanding of the process of ozone formation. A lucid and regular seasonal variation was observed in the study area for the entire one-year period of measurement. The seasonal mean of diurnal values of SOZ concentration for the two sites are given in Table 1 and depicted in Figs 4(a-d). The summer days are with high solar flux, high temperature and clear skies with less rainfall. These factors make summer season to produce more SOZ concentration. Similar seasonal trend with different values were observed at both sites. Southwest monsoon recorded the second highest concentration of SOZ concentration. Thick cloud coverage and heavy rainfall caused low levels of SOZ during northeast monsoon. Relatively low levels of ozone during the winter as compared to summer might be due to greater atmospheric stability and an increased incidence of nocturnal inversions[29].The formation of ozone in the troposphere depends upon several meteorological factors, such as temperature, sunshine hours, wind velocity and relative humidity[30]. A similar seasonal dependence of SOZ has been reported in several studies conducted in USA[31,32], Europe[33], Canada[34] and Korea[35].

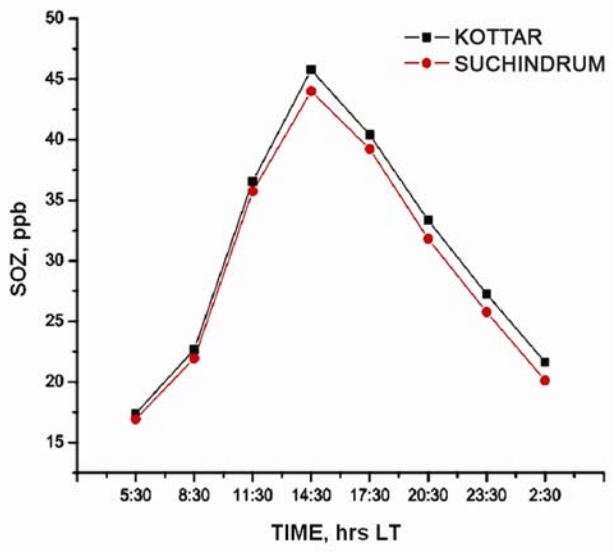

Fig. 3 — Diurnal variation of SOZ

4.3 Diurnal rate of change

The rate of change of SOZ concentration for the overall seasonal diurnal values was calculated and is shown in Fig. 5. The rate of change is helpful in understanding the atmospheric potential to buildup SOZ concentration within regular intervals of time. The overall rate of change at 0830 and 1130 hrs LT during the entire period of study for the two sites was found to be 4.62 and 4.59 ppb, respectively and at 1730 and 2030 hrs LT, it was found to be around -2.02. These values are well comparable with other studies. Elampari *et al.*[16] showed that the rate of change at 0830 and 1130 hrs LT in Nagercoil was around 4.2 ppb. Also, the maximum rate of change in Johrapur and Delhi were found as 4.5 ppb and in Gadanki, it was observed as 4.6 ppb. It is evident that irrespective of the vehicular emissions, there are some other unclear factors like surface meteorology,

Table 1 — Seasonal mean diurnal values of SOZ concentration

Seasons	Average O_3 concentration (ppb) at different times (hrs LT)							
	05:30	08:30	11:30	14:30	17:30	20:30	23:30	02:30
Site1 (Kottar)								
Summer	19.04	26.29	42.43	50.83	44.96	37.87	31.94	24.40
Southwest monsoon	16.99	22.26	35.98	45.68	40.59	34.21	27.52	21.85
Northeast monsoon	15.26	19.18	31.29	44.55	34.90	27.62	22.54	22.24
Winter	19.00	23.32	36.81	46.08	41.48	33.45	26.84	18.21
Site2 (Suchindrum)								
Summer	18.59	25.42	41.46	48.52	42.14	34.30	29.44	21.79
Southwest monsoon	16.94	21.62	35.39	44.50	39.90	33.59	25.89	20.22
Northeast monsoon	14.48	18.78	30.91	39.31	34.70	26.92	21.73	17.73
Winter	18.17	22.22	35.08	43.24	40.41	31.94	26.14	21.09

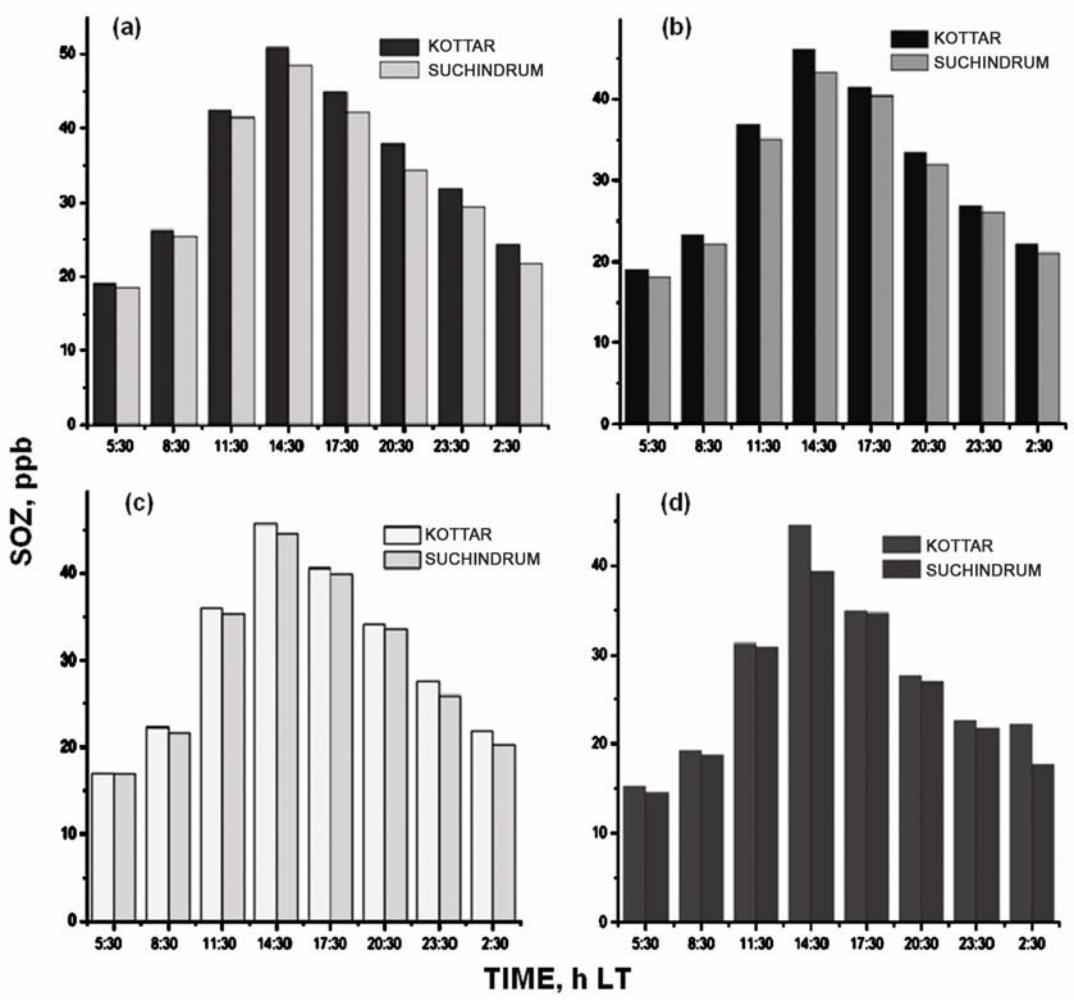

Fig. 4 — Seasonal variation of ozone during: (a) summer, (b) winter, (c) southwest monsoon, and (d) northeast monsoon

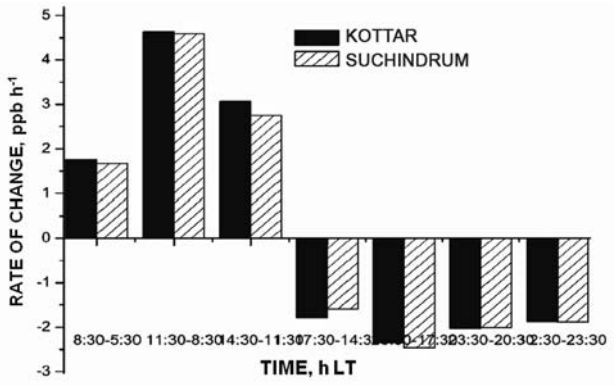

Fig. 5 — Rate of change of ozone

boundary layer and VOC to NOx ratio that decides the variation of SOZ at a particular place.

4.4 SOZ and Surface meteorology

The production of SOZ is controlled by temperature, sunlight, humidity and the long range transport of pollutants all of which are sensitive to changes in climate. Climate change is inherently coupled to changes in regional and local meteorology, which may affect the potential for build-up of harmful levels of SOZ in polluted areas. Transport of pollutants and their resulting concentrations depend on meteorological parameters such as temperature, wind speed, direction, atmospheric stability and the movement of the pressure systems[36]. The meteorological conditions that help the formation of ozone are intense solar radiation, low wind speed, high temperature and a restricted boundary layer depth[37]. Clear skies, warm temperature, solar radiation and soft winds are believed to have a great influence on surface ozone concentration[38]. The mean temperature of air at any place depends on various factors, of which, altitude, latitude, proximity to the sea, temperature of the sea and exposure are the main ones[39]. The ozone concentration reaches its peak

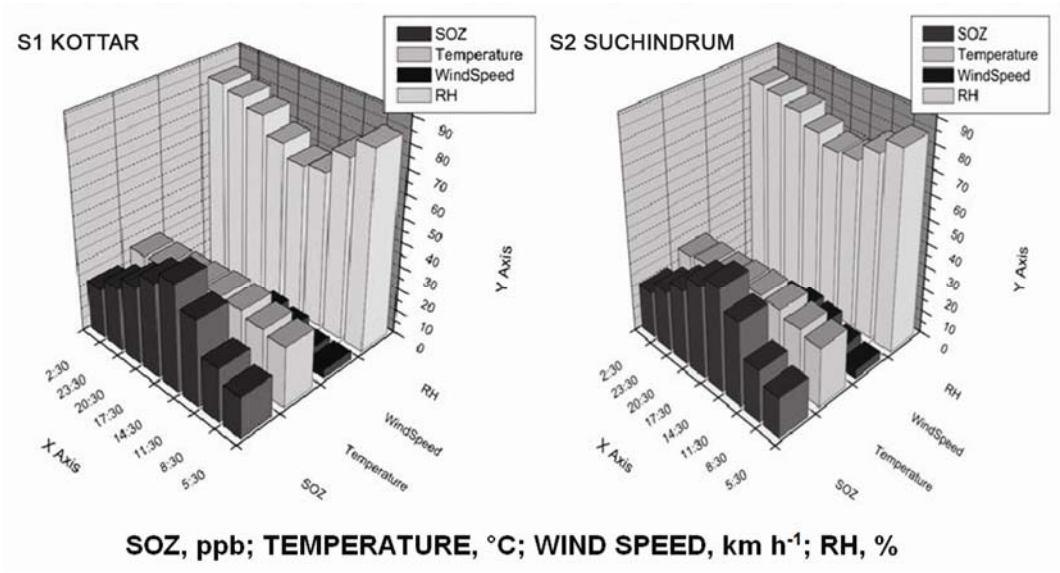

Fig. 6 — Variation of SOZ with meteorology at two sites

Table 2 — Correlation of surface ozone with various parameters

Meteorology	Correlation coefficient (*r*)	
	S1 (Kottar)	S2 (Suchindrum)
Temperature	0.712788	0.70763
Wind speed	0.323673	0.425224
RH	-0.5369	-0.4936

Table 3 — Descriptive statistics of the daily average surface ozone at two sites

Statistics	Site 1	Site 2
Sample size	109	109
Maximum	56	52
Minimum	13	12
Mean	31.1357	29.1239
Standard deviation	3.4570	3.0556

value when the temperature is the maximum, which indicates that ozone concentration levels are directly related to temperature. Ambient air temperatures differ with seasons of the year and time of the day. In all seasons, it was observed that the highest temperature reached at 1430 hrs LT.

The overall correlation between temperature and SOZ concentration for the entire study period was found to be positive (r = +0.712788, p = 4.314E-05) for site S1 and (r=+0.707639, p = 6.2134E-05) for site S2, respectively. The variation of SOZ with temperature, RH and wind speed is shown in Fig. 6. Table 2 shows the correlation of surface ozone with the meteorological factors. The descriptive statistics of the surface ozone data obtained from the two sites is shown in the Table 3. From the data points

Table 4 — t-test results

Parameter	Value
t-value	4.5525
t-critical	1.9710
h	1
p	8.8506e-006
Ci	1.1408, 2.8829
df	216

obtained, it is evident that both the sites are characterized by almost similar conditions of surface meteorology. Even though, both the sites were favoured by almost same meteorological conditions, a difference of 2.01 ppb was observed in the mean of daily average surface ozone concentration between the two sites. Statistical two tailed t-test was performed to trace out this significant difference by framing the null hypothesis as 'for similar meteorological conditions, the surface ozone concentration remains the same'. Table 4 shows the results of t-test with 5% significance level on the surface ozone data for the two sites. From the test results, since h=1 and t-value calculated from the data is now larger than the critical value t-critical, the null hypothesis can be rejected and it is concluded that the means are not identical at a 5% significance level. The p-value is very close to zero, suggesting that the null hypothesis is very unlikely to be true. The 95% confidence interval on the mean is [3.9742, 2.1702], which again includes the difference between the mean as 2.01 ppb. Thus, it confirms that the difference in the surface ozone concentration at both the sites influenced by same meteorological

conditions is only because of the difference in the levels of the VOC and precursor gas.

5 Conclusions

Surface ozone measurements were made at two sites Kottar (8.1739°N, 77.4389°E) and Suchindrum (8.1550°N, 77.4650°E) of Kanyakumari district, Tamil Nadu, India for a period of one year from February 2014 to January 2015. Eight readings were taken for all possible days starting from 0530 hrs LT in the early morning to next day 0230 hrs LT with an interval of three hours. The two sites were chosen in such a way that Kottar region is busy with commercial activities and more polluted than Suchindrum. The two regions are influenced by almost similar meteorological conditions since they are not far away from each other. Aeroqual monitor with various sensor head was used for measuring surface ozone. Meteorological parameters were also recorded by the similar instrument. In addition, the meteorological readings were obtained from Central Water Commission Department, Asramam, Suchindrum. The readings were also obtained from IMD website. Surface ozone at both the sites showed a clear diurnal pattern. Diurnal variation tends to be highly related to sunlight intensity throughout the day. As a result, during early morning hours, when the sun starts to shine, the ozone concentration started increasing. Moving towards mid-day, the sunlight being strongest, resulting in high ozone and then the concentration decreased in late evening hours. The concentration of surface ozone at the both the sites are within the air quality standards (60 ppb). The site S2 (Suchindrum) recorded less amount of surface ozone as compared to the site S1 (Kottar). The maximum value of SOZ recorded was 56 ppb. Summer months recorded maximum ozone followed by south west monsoon and winter. Low levels of ozone were recorded during northeast monsoon. The rate of change of SOZ concentration was very well compared with other places. Statistical test was performed to confirm the significance of difference in mean values between two sites. Even though they have similar surface meteorology, the results show that the difference is because of the difference in the precursor emission levels.

Acknowledgement

The authors acknowledge CWCD, Asramam, IMD Kanyakumari and weather underground website for supporting meteorological data.

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