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Daytime Ozone Variation in Surface Air in a Subtropical Mangrove Estuary at Manakudy, South India

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ABSTRACT: Surface ozone (SOZ) can be very harmful, if it exceeds the threshold limit. It can accumulate over sea and return to the land along with the breeze. Rural and vegetation rich areas often record elevated levels of surface ozone, due to the variations in Volatile Organic Compound (VOC) levels as well as wind velocity and direction. Similarly, methane is also an important greenhouse gas, which plays a vital role in atmospheric budget. The current study analyzes ozone and methane levels, measured during daytime in a mangrove estuary near Manakudy (8.0911 N, 77.477 E), Kanyakumari, South India for a period of ten months, from March 2014 to December 2014. SOZ shows an imprecise diurnal pattern with an early morning peak, whereas methane records an apparent diurnality. The maximum value of SOZ is around 50 ppb, with Summer months possessing high levels of SOZ, followed by Southwest monsoon (SWM) and Northeast monsoon (NEM). High levels of methane are found in SWM followed by NEM with the lowest concentration occurring in Summer. The correlation between SOZ and methane is found to be $r = -0.257$, $p<0.001$. The positive correlation of SOZ with temperature and wind speed strongly suggests the role of such surface meteorology in SOZ production and transport.

Keywords: VOC, estuary, diurnal, greenhouse, correlation, meteorology

INTRODUCTION

In the troposphere, ozone has elevated as a result of increased human-produced ozone precursor emissions. Ground level ozone is responsible for reduced crop production in the United States each year (EPA US, 2003) through its interference with a plant's capacity to produce and store food, thereby increasing its vulnerability to disease, insects, other pollutants, and harsh weather (EPA US, 1999b). In humans, elevated ozone levels impair lung function, irritating the respiratory tract by damaging the bronchioles and alveoli, which leads to permanent damage with repeated exposure. Even at lower concentrations, breathing ozone for sustained periods can initiate a variety of health issues such as chest pain, cough, nausea, throat irritation, and congestion, while exacerbating existing cases of bronchitis, heart disease, emphysema, and asthma. Climate changes may also affect meteorological transport processes, thus altering the export and import of pollution (Doherty et al.,2013). Methane is the primary component of

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natural gas. It is present under the earth's surface in vast quantities, though its levels in the atmosphere are relatively low. Methane is produced naturally by volcanoes, ruminant animals such as cattle and sheep, decaying plants, extraction of natural gas, coal mining, and waste disposal methods like landfills. A major 'greenhouse gas', methane can be released into the environment during its extraction from the earth, emissions from industries, agriculture, distribution, and use in residential and commercial buildings. Although its photo-oxidation is slow on the timescale of urban air pollution, Crutzen (1973) pointed out that on the timescale of air masses in the global troposphere, reactions by methane are a significant source of tropospheric ozone, comparable to or even greater than ozone transport to the troposphere from the stratosphere.

Ozone is present in the natural, unpolluted troposphere, the density of its tropospheric column being approximately 10% of the total atmospheric (i.e., troposphere plus stratosphere) ozone column density (Logan et al., 1981; Brühl and Crutzen, 1989; Fishman et al., 1990). The ozone in the stratosphere absorbs short-wavelength radiation ($\lambda \leq 290$ nm) from the sun, only allowing them to penetrate into the troposphere (Peterson, 1976; Demerjian et al., 1980). It is recognized that trace gases like NO, NO_2, CO, VOC's, and NMHCs are the precursors of surface ozone. In the unpolluted atmosphere, ozone can be formed through photolysis of oxygen by UV radiation with a wavelength of 240 nm or less (McElroy, 2002) in the presence of a third molecule such as nitrogen that helps absorbing the energy, released by photolysis. The knowledge of surface ozone mole fractions and their global distribution is of utmost importance due to the impact of ozone on human health and ecosystems, not to mention the central role of ozone in controlling the oxidation capacity of the troposphere (Anet et al.,2017). Major emission sources of these air pollutants are anthropogenic activities like fossil fuel combustion and biomass burning (Ohara et al., 2007; Streets et al., 2013). Tropospheric ozone is formed from the photochemical reaction of NO and hydrocarbons, that is why it is referred to as a secondary pollutant since there are no significant direct, man-made (anthropogenic) emissions of ozone into the troposphere. NOx and VOC's are emitted through natural processes, such as bacterial activity in soil, forest fires, and VOC release from vegetation. In the troposphere, nitrogen dioxide is the only known compound that can produce O atom during its photodissociation with available radiation. The main objective of this work is to study day-time variation of ozone and methane alongside meteorology of a tropical estuary, Manakudy of Kanyakumari District, India.

MATERIALS AND METHODS

The measurements took place in Manakudy estuary in Kanyamari district, one of the smallest districts in Tamil Nadu with total geographical area of 17,672 sq.Km. It is bound by Tirunelveli District on the North and East, the Gulf of Mannar on the Southeast, the Indian Ocean and the Arabian Sea on the South and Southwest, and Kerala on the West and Northwest. The District has a favourable agro-climatic condition, making it suitable for growing a number of crops. According to Assistant Director of Statistics, Nagercoil, this region's climate is divided into four seasons: The winter extends from January to February, followed by summer from March to May. The Southwest monsoon (SWM) period starts from June and ends in September, while the Northeast monsoon (NEM) period begins from the month of October and ends in mid-December. On the whole, this area experiences a tropical type of climate. Unlike other districts in Tamil

Nadu State, Kanyakumari District experiences rainfall during both Southwest and Northeast monsoons, displaying a distinct variation in the climatic conditions themselves. As such, the proximity of the equator, its topography, and other climate factors favour the growth of various crops.

SOZ and methane measurements were carried for the first time in and around a mangrove estuary at Manakudy (4 km from Kanyakumari). Mangrove swamps are found in tropical and subtropical tidal areas. Areas where mangal occurs include estuaries and marine shorelines. Figure 1 shows the study area. Methane emissions from estuarine surfaces vary over a wide range of spatial and temporal scales. The process by which this gas is produced, transported, and emitted cannot be clearly explained as it is very complex. The behavior of this gas differs in tidal flats, wetlands, and marshes, compared to estuaries, for methane production is intense in tidal flats and marshes.

Day time readings were recorded. They were taken on possible days for ten months, from March 2014 to December 2014, which comprises three seasons and the measurements were taken using Aeroqual S 200 and S 500 monitor with datta logging facility. Surface ozone and methane concentrations in the ambient air were measured daily at every 3 hours from 05:30 to 17:30. Temperature and wind speed are the key meteorological parameters, measured at the site.

Fig.1. Study Area (Manakudy Mangrove Estuary)

Fig. 2. Aeroqual Monitor

The small size and portability of the instrument allowed it to be deployed in strategic locations within the estuarine system. In the sampling sites, the monitor was positioned at least 100 m away from the wetland in order to minimize perturbations from estuarine wetlands.

RESULTS AND DISCUSSION

Previous studies of O_3 concentrations have indicated considerable variability from day to day and year to year, as a result of not only changes in precursor emissions, but also meteorology (Agudelo-Castaneda & Teixeira, 2013). Diurnal ozone variation is an atmospheric term that relates the variation in the surface ozone, occurring from the high rates of the day to the low rates of the night. In general it stands for ozone fluctuations, occurring each day. It is very useful in understanding the various processes, responsible for the production and destruction of ozone, being a measure for overall budget of production and loss rates. The diurnal variation is characterized by a considerable minimum rate at night, a rapid ascent in the morning after sunrise, and an absolute maximum near noontime (Widen, 1966). In general, SOZ concentration show a clear diurnal pattern, characterized by minimum values around 05:30 in early morning, a gradual build-up after 08:30, and a maximum rate in the afternoon at 14:30. However, in the study area, a peak was obtained around 05:30, which was quite different from other area measurements such as roadside, polluted zones, and the value decreased at 0830 am. Figure 3 illustrates the day time variations of SOZ, showing a gradual increase with the maximum peak recorded at 14:30. Due to the lower boundary layer height, mainly reducing the mixing process between ozone-low surface layer and ozone-rich upper layer, there was a generally low concentration in the morning hours. The peak at around 05:30 could be due to the accumulation of previous day's SOZ as well as the lack of midnight NOx titration. The mixing ratios of ozone start increasing gradually after sunrise, attaining maximum values during local noon time (Nishanth & Satheesh Kumar, 2011). Daytime higher ozone levels were mainly due to photochemical production of ozone (Naja & Lal, 2002). Apart from the role of photochemistry, both boundary layer meteorology and dynamics play a key role in ozone variability, as well (White & Templeman, 2002). Boundary layer attained the maximum height during afternoon hours, as a result of the increase in surface heating. During this time, trace species got vigorously mixed within, forming convective mixed layers (Reddy & Rama Gopal, 2008). From the diurnal variations, it was evident that ozone concentration tended to be highly related to the amount of sunlight throughout the day. During early morning hours, when the sun starts to rise, the ozone production rate started increasing, since the sunlight helped increasing ozone production rate. Moving towards mid-day, when the sunlight was the strongest, resulted in high ozone production. Increased ozone concentration in daylight can be attributed to photolysis reactions of NO_2 and photo oxidation of VOCs, CO, and hydrocarbons. It also could be attributed to downward transport of ozone via vertical mixing (Tyson et al.,1998).

The minimum value of averaged diurnal CH_4 Concentration was found to be 1.77 ppm and the maximum one, 4.73 ppm. Many studies reveal that diurnal variations of methane is not clear because not only does the concentration depend on anthropogenic sources and bio mass burning, but factors like soil temperature and different stages of crop growth (e.g. tillering, panicle initiation, booting, flowering and maturity) play an important role, also. Figure 4 shows daytime variations of methane, according to which there was an apparent diurnal pattern in

methane with two distinct peaks: one early in the morning and the other around 17:30 in the evening. The sunrise peak was consistent with two-way transport mechanism, i.e., convective in the daytime and diffusive in the night-time. Afterwards, the methane concentration dropped until 14:30. The low values occurred between 14:30 and 23:30, while the maximum peak was observed around 17:30 in the evening. Due to the fact that the larger drop in atmospheric pressure took place in late morning rather than at night, the evening peak was higher than the early-morning peak (Chandrasekhar & Balaji, 2012). This type of double-peak is often seen in "urban pollutants" such as carbon monoxide, where morning and evening rush hours lead to elevated mole fractions (Panday & Prinn, 2009).

The seasonal variation is important for better understanding the process of ozone formation. Table 1 presents the monthly mean values of SOZ, showing that summer months recorded the highest concentrations of SOZ, followed by Southwest monsoon and winter. The summer reading corresponded to the averaged values of SOZ for three months of March, April, and May. Furthermore, low levels of SOZ concentration were recorded during northeast monsoon, which were because of more cloudy days, less sunshine hours, and insufficient solar flux, along with pollutants' washout as a consequence of heavy rainfall.

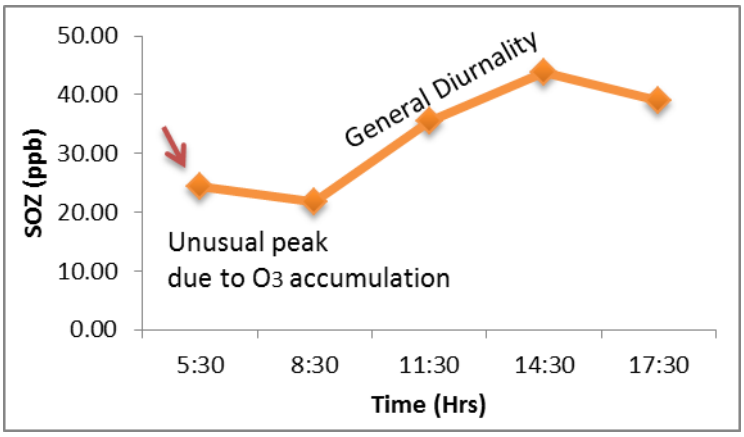

Fig. 3. Daytime variations of SOZ

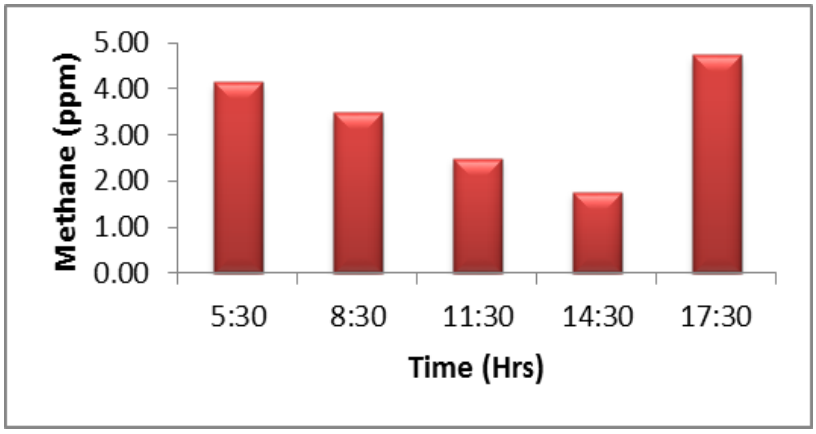

Fig. 4. Daytime variations of methane

Table 1. Monthly average values of SOZ (ppb)

Month/Time	5:30	8:30	11:30	14:30	17:30
March	20.16	26.03	40.57	47.35	42.20
April	27.00	25.06	40.41	49.63	43.57
May	21.00	25.57	41.37	49.87	45.45
June	23.80	24.29	39.32	48.81	44.60
July	23.00	22.45	36.75	45.25	40.26
August	24.40	20.80	34.25	42.70	37.96
September	24.00	20.08	32.20	40.71	35.40
October	25.00	17.60	30.14	38.89	33.88
November	26.38	17.19	28.02	35.45	31.53
December	26.90	19.53	31.76	38.75	34.69

The summer days are characterized by high solar flux, high temperature, and clear skies, along with less rainfall. These factors make summer season produce more SOZ concentration. Southwest monsoon recorded the second highest concentration of SOZ concentration. Figure 5 illustrates the seasonal variations of SOZ. Even though the district receives rain from both monsoons, the southwest one is characterized by less rainfall, when compared to that of northeast monsoon. Concerning the northeast monsoon, the morning values were around 26 ppb, while the maximum values ranged around 38 ppb during noontime. Thick cloud coverage and heavy rainfall caused low levels of SOZ in this season. Even though there were vehicular emissions during this season, there was no sufficient solar flux density to convert the precursors to SOZ concentration. Also, the rain washed away these pollutants without allowing them to settle down.

Emission of methane into the atmosphere is directly related to temperature and moisture. Thus, natural environmental changes that occur during the seasonal change act as a major control for methane emission. Additionally, even the changes in temperature during the day can affect the amount of methane, produced and consumed. For example, methane-producing plants can emit as much as two to four times more methane during the day than the night, which is directly related to the fact that plants tend to rely on solar energy to enact chemical processes. Additionally, methane emissions are affected by the level of water sources. Seasonal floods in the spring and summer naturally increase the amount of methane, released into the air. Table 2 presents the monthly mean values of methane.

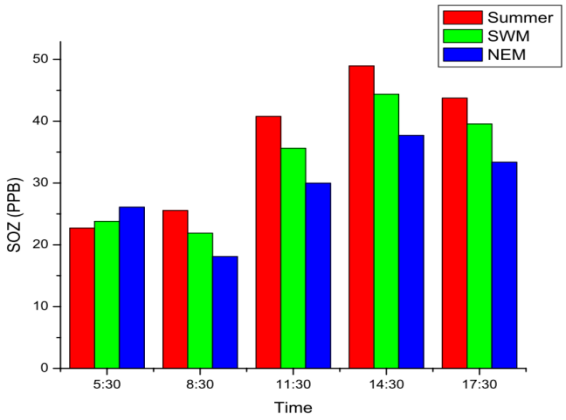

Fig. 5. Seasonal variations of SOZ

Table 2. Monthly average of Methane (ppm)

Month/Time	5:30	8:30	11:30	14:30	17:30
March	2.33	2.33	1.50	1.33	4.83
April	2.00	2.17	1.17	1.00	5.17
May	2.33	2.00	1.00	0.83	3.83
June	5.67	4.33	3.17	1.83	4.83
July	6.50	4.00	3.33	2.33	5.17
August	6.33	4.17	3.00	2.17	4.50
September	6.67	4.50	3.33	2.50	5.33
October	3.33	3.33	2.67	2.00	4.50
November	2.83	4.33	3.00	2.33	4.83
December	4.50	3.50	2.67	1.83	4.50

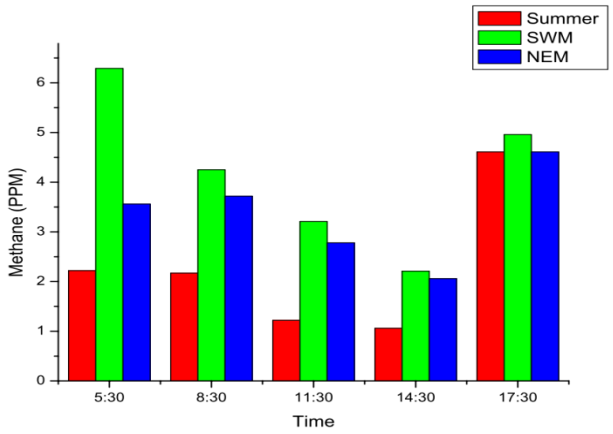

Fig. 6. Seasonal variations of Methane

SWM (June-September) recorded the highest concentration of methane, followed by NEM, while the low levels were observed in summer. The peak in CH_4 emissions occurred during the wet season (June to September) when the majority of rice in South Asia is generally grown (Pathak & Wassmann, 2005). Summer months never showed any distinct peak in early morning hours. Results suggested that the pollutants, strongly dependent on vehicular emissions, had higher peaks during summer months.

The rate of SOZ change and Methane concentration for the overall seasonal diurnal values was calculated and can be seen in Figure 7.

$$\text{Rate of change of ozone } \left[\mathrm{d}\left(\mathrm{O}_3\right)/\mathrm{dt}\right] = \frac{O3(8.30\ hrs) - O3(5.30\ hrs)}{3}$$

The overall rate of change for SOZ between 08:30 and 11:30 during the entire period of study was 4.55 ppb and – 1.58 ppb between 14:30 and 17:30. The reason behind this is primarily due to the intensity of solar radiation and emission levels. For methane, the rate of change was observed as 0.98 ppm between 14:30 and 17:30. A high positive value of rate of indicates a sudden increase in the pollutant levels between the given interval of time and vice versa.

The mean temperature of the air at any place depends on many factors, of which, altitude, latitude, proximity to the sea, temperature of the sea, and exposure are the chief (Rao & Ramamurthy, 1972). Higher temperatures may also result in enhanced local production of O_3 by boosting HOx production or increasing local biogenic or

fugitive anthropogenic (such as the evaporation of oil and/or gas) VOC emissions (Pugilese et al., 2014). It is well known that SOZ is highly variable and the trends, derived in one location, may not represent the whole region. Many factors exist simultaneously at different times or locations, rendering the real system much complicated; however, several studies suggest that climate impacts and –especially-- temperatures are strong enough to affect tropospheric ozone distribution (Hsu, 2007). Temperature and long-term urban warming have a serious impact on urban pollution, contributing to higher ozone concentrations, as the heat accelerates the chemical reactions in the atmosphere (Walcek & Yuan, 1999). Figure 8 demonstrates the diurnal temperature variation, showing that the ozone concentration had reached its peak value when the temperature was the maximum, indicating that ozone concentration levels were directly related to temperature. Ambient air temperatures differ with seasons of the year as well as the time of the day. In all seasons, it was observed that the highest temperature was reached at 14:30.

The overall correlation between temperature and SOZ concentration for the entire study period was found to be positive **(**r = +0.63, p =9.37E-54). The correlation between temperature and CH_4 within the study area was found to be as the following: r =-.310, P=4.84E-12. For methane, higher concentrations occurred during late evening and early morning hours around sunrise. Since methane gas is not primarily emitted from vehicle exhausts, it is not involved in temperature-dependent photochemical activity as SOZ. Tables 3 and 4 present the correlation of temperature with ozone and methane is given in respectively.

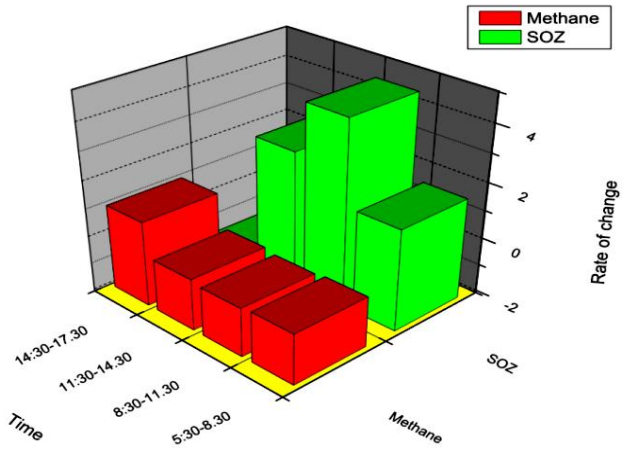

Fig. 7. Rate of change

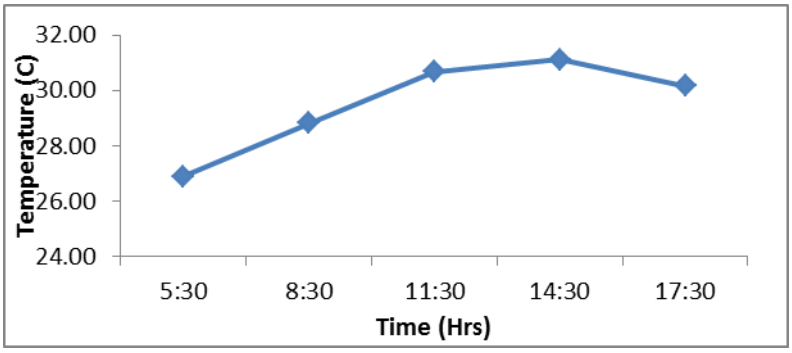

Fig. 8. Daytime variations of the temperature

Table 3. Correlation between temperature and Ozone

		Temperature	Ozone
Temperature	Pearson Correlation	1	.634**
	Sig. (2-tailed)		.000
	N	465	465
Ozone	Pearson Correlation	.634**	1
	Sig. (2-tailed)	.000	
	N	465	465

** Correlation is significant at the 0.01 level (2-tailed).

Table 4. Correlations between temperature and methane

		Temperature	Methane
Temperature	Pearson Correlation	1	-.310**
	Sig. (2-tailed)		.000
	N	465	465
Methane	Pearson Correlation	-.310**	1
	Sig. (2-tailed)	.000	
	N	465	465

** Correlation is significant at the 0.01 level (2-tailed).

Table 5. Correlation between methane and ozone

		Methane	Ozone
Methane	Pearson Correlation	1	-.257**
	Sig. (2-tailed)		.000
	Sum of Squares and Cross-products	1812.895	-2323.662
	Covariance	3.907	-5.008
	N	465	465
Ozone	Pearson Correlation	-.257**	1
	Sig. (2-tailed)	.000	
	Sum of Squares and Cross-products	-2323.662	44985.117
	Covariance	-5.008	96.951
	N	465	465

** Correlation is significant at the 0.01 level (2-tailed).

In many ozone-monitoring stations, there is a positive correlation between hourly wind speed and hourly ozone concentration. The overall correlation between SOZ and wind speed in the study area is positive ($r = +0.40$, $P < 0.0001$). Wind speed and wind direction are the two important parameters for dispersion and transport of ozone and its precursors from their emission sources. Wind direction controls the way of pollutant dispersion, while wind speed decides the distance of the dispersion. Wind and ozone both vary on a similar diurnal pattern, with higher wind speeds and higher ozone concentrations usually being observed during daytime. At certain times of year, high wind speed (or the synoptic weather conditions that go along with it) may be effective at mixing higher concentrations of ozone to the surface from the free troposphere. The above factors will lead to a positive correlation between wind speed and ground level ozone. Light winds reduce atmospheric dispersion, allowing greater concentrations of precursor pollutants to accumulate in the boundary layer, which in turn leads to greater ozone formation. During the entire period of the study, the wind speed varied between 2 km/h and 27 km/h. The correlation between wind speed and CH_4 was also

weak in the study area (r = - 0.1064, P<0.005). The correlation between ozone and methane in the study area was found and is given in Table 5.

CONCLUSION

Surface ozone (SOZ) is a major component of photochemical smog, both a known human health hazard and a damaging factor for vegetation. Its precursor compounds, namely, nitrogen oxides (NOx) and volatile organic compounds (VOCs), have a variety of anthropogenic and biogenic sources, exhibiting non-linear effects on ozone production. SOZ and Methane measurements were carried out in a mangrove estuary at Manakudy, Kanyakumari District, South India, for ten months from March 2014 to December 2014. Diurnal patterns of ozone at the study area tended to remain highly related to sunlight intensity throughout the day with the help of precursors, generated from human activities. SOZ showed an imprecise diurnal pattern with an early morning and afternoon peaks, whereas methane recorded an apparent diurnality. The maximum value of SOZ was around 50 ppb. The early morning peak, obtained around 05:30, was pretty different from other area measurements, like roadside and polluted zones. Many studies account for low concentration of SOZ during early morning. The main reason for high values in the study area may be due to the absence of NOX titrations, followed by the accumulation of SOZ. There was an apparent diurnal pattern in methane with two distinct peaks: one during early morning and the other around 17:30 Lt in the evening. Summer months recorded the highest concentration of SOZ followed by Southwest monsoon and winter. Low levels of SOZ concentration were recorded during northeast monsoon. SWM (June-September) recorded the highest concentration of methane, followed by NEM. The low levels were observed in summer. This factor had a significant positive correlation with temperature and wind speed and as the study clearly showed, the estuarine meteorology had had an obvious impact on greenhouse gases. Results revealed that the surface ozone concentration in the study area was well within the desirable limits, yet ozone showed some potentiality to increase in near future. As a result, continuous monitoring and analysis is very much essential to frame mitigation strategies. Future studies should also be directed at elaborating the effect of other meteorological parameters such as dew point, wind direction, and the inversion layer height, since it was indicated that the latter does play a role in the ozone and its precursor concentrations, as well.

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