

# Seasonal variation of surface ozone and its association with meteorological parameters, UV-radiation, rainfall and cloud cover over Chennai, India

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A study was done to measure the surface ozone ( $O_3$ ) level during the period between June 2011 and September 2012 at Chennai, a tropical site on the south-east coast of India situated at 13°04'N 80°7'E. Surface ozone and its variation with meteorological parameters in the urban area of Chennai were analysed through temporal pattern. The seasonal ozone showed a unique pattern attaining peak levels in summer and weakening in winter. Higher surface ozone concentrations observed during the summer months of the present study can be attributed to the high intensity of solar radiation and high temperature levels which promote the photochemical generation of  $O_3$ . The destruction of  $O_3$  is increased during the winter season as a result of scavenging of higher nitrogen oxides. Correlations were observed and analysed to understand as to which meteorological variables influence the formation of ozone the most. Finally, hourly instantaneous values measured during daytime varied from 10 to 40 ppbv and its day average varied from 13 to 30 ppbv.

**Keywords:** Diurnal cycle, meteorological parameters, radiative forcing, seasonal variation, surface ozone.

OZONE ( $O_3$ ) is a protective component in the stratosphere. It absorbs and prevents the major ultraviolet (UV)-rays of solar radiation with wavelength less than 280 nm (UV-C) from reaching the Earth's surface<sup>1</sup>. However, ozone is considered as a pollutant at the ground level and the overall effect of ozone exposure leads to decrease in lung capability to perform normal functions<sup>2</sup>. Hence, ozone can be regarded as a beneficial UV shield in the stratosphere, but harmful to human beings at the ground level. An increase in ozone might contribute to a warming of the Earth's atmosphere<sup>3</sup>. Ozone absorbs the Earth's infrared radiation at 9.6  $\mu\text{m}$  and contributes to the greenhouse effect. The radiative forcing (defined as the difference between radiant energy received by the Earth and energy radiated back to space in  $\text{W}/\text{m}^2$ ) of ozone per molecule

basis is 1200–2000 times that of  $\text{CO}_2$  and is much more than that of methane ( $\text{CH}_4$ ) and  $\text{N}_2\text{O}$  (refs 4 and 5). The most important objective of air quality policy for Governments all over the world is the reduction in surface ozone, which is harmful to human, animal and plant health<sup>1</sup>.

Furthermore, it was observed from laboratory experiments that a large amount of flammable materials in fire-works produce large amounts of ozone. In addition to emitting light in the visible region, metals at high temperature also emit radiation in the UV region. Consequently, the high-energy UV-radiation is absorbed by molecular oxygen present in the air and this results in the splitting of molecular oxygen into atomic oxygen. This in turn reacts with molecular oxygen to produce ozone<sup>6</sup>. Although surface ozone exhibits high natural variability throughout the year, there will be short-term enhancement in ozone levels coupled with high levels of pollutants in the environment during festival seasons. The ambient air quality standards formulated and adopted by the United States Environment Production Agency (USEPA) accepts 120 ppb of ozone for 1-h average and 80 ppb for 8-h average<sup>6</sup>.

Ozone formation is an endothermic reaction and so higher temperature will favour higher equilibrium concentration of ozone<sup>7</sup>. A large number of observations have shown that the concentrations of ozone will depend on intensity of solar radiation and temperature. In addition, the magnitude of ozone variation is high on clear days than cloudy or winter days<sup>8</sup>. The weekend effect of ozone – a phenomenon with high ozone concentration during weekends compared to weekdays, and relatively low concentration of precursors ( $\text{NO}_x$  and volatile organic compounds (VOCs)) at weekends – has been reported in some areas of America<sup>9,10</sup> since 1970 and Japan<sup>11</sup>.

## Basic ozone chemistry

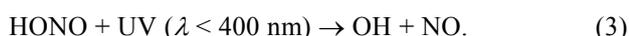
VOCs have a direct impact on the environment, but contribute indirectly via secondary organic aerosol (SOA) formation in climate change. In urban and industrialized

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regions, use of fossil fuel is a significant source of VOCs. The oxidation efficiency of VOCs is determined by the reactions with hydroxyl (OH) radicals, O<sub>3</sub> and nitrate (NO<sub>3</sub>) in the atmosphere. The OH radicals are formed through the photo-dissociation of O<sub>3</sub> by solar UV radiation<sup>12</sup>.



The photolysis of nitrous acid (HONO) can also be a significant source of OH in highly polluted regions.



Though OH radicals react with many trace constituents, the oxidation rates of VOCs are much faster than the relatively more abundant trace gases like CH<sub>4</sub> and carbon monoxide (CO). The following nonlinear reactions involving VOCs and NO<sub>x</sub> in the presence of solar radiation lead to the production of O<sub>3</sub> and a variety of partially oxidized species. The following equation is a simplified version of many complex chemical reactions, and VOCs react with OH radical to finally produce O<sub>3</sub>.



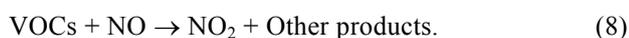
where CARB represents carbonyl compounds in the atmosphere<sup>12</sup>. The Earth's troposphere acts like a chemical chamber in which huge quantities of trace species are transformed from one form to another, where ozone can be considered as the principal product of this reaction. Most of the OH radicals are produced by ozone during daytime. In the presence of UV-radiation ( $h\nu$ ), oxygen (O<sub>2</sub>) and nitrogen dioxide (NO<sub>2</sub>) react in the atmosphere to form ozone and nitric oxide (NO) through the reactions given in eqs (5) and (6).



The resultant ozone, however, is quickly reacted away to form nitrogen dioxide by the process given in eq. (7). This conversion of ozone by NO is referred to as titration. In the absence of other species, a steady state is achieved through the reactions shown by eqs (5), through (7)<sup>13</sup>.



Ozone cannot accumulate further unless VOCs, which include hydrocarbons, are present to consume or convert NO back to NO<sub>2</sub>, as shown in eq. (8).



This equation is a simplified version of many complex chemical reactions. As NO is consumed by this process, it is no longer available to titrate ozone. When additional VOCs are added to the atmosphere, a greater proportion of the NO is oxidized to NO<sub>2</sub>, resulting in greater ozone formation. The rate of photochemical production of O<sub>3</sub> in the urbanized/polluted regions not only depends on the ambient levels of VOCs and NO<sub>x</sub>, but also on their ratio (VOC/NO<sub>x</sub>). Because of this complexity (or nonlinearity) in the chemistry and the roles of various meteorological parameters, the quantitative links of emissions of VOCs and NO<sub>x</sub> to the concentrations of O<sub>3</sub> and major photochemical oxidants at a particular location and time are not straight forward<sup>13</sup>.

### Objective of the study

Surface measurement of ozone and some of precursors is being done at several sites in India, but not on a regular basis. According to model calculations, there will be a large increase in surface ozone in the Asian region by the end of this century due to increased emission of ozone precursor gases<sup>14</sup>. So more concentrated measurements are needed to describe natural variation and changes due to anthropogenic sources over the highly polluted urbanized Indian region. In this study, an attempt has been made to address briefly some of the important issues, relevant to the changing climate scenario, with special emphasis on temporal (diurnal and seasonal) variations in surface ozone over a tropical urban site. A study on the interrelation of ozone with available meteorological parameters, viz. relative humidity, temperature, rainfall, wind speed, UV-radiation and cloud cover is carried out and discussed.

### Study area

Chennai, which is situated on the southeast coast of India and northeast coast of Tamil Nadu, has a 19 km long shoreline along the Coromandel coast on a flat coastal plain known as the Eastern Coastal Plains. This area is one of the most highly populated urban sites. Chennai lies on the thermal equator and is also a coastal city. The latitude and longitude of the centre of the city are 80°14'51"E and 13°03'40"N. The city area is about 70 sq. miles and the metro (overall) area is about 456 sq. miles. The elevation above sea level is 6 m and this prevents extreme variation in seasonal temperature. The geographical location of the experimental site is shown in Figure 1; it is located in south Chennai. The different sources of air pollution in Chennai city are classified under the following categories – transport, industries, residential and others. Chennai can be divided into four areas – north, central, south and west. The northern part is primarily an industrial



urban areas of India<sup>23</sup>. To the best of our knowledge, no measurements have been carried out over Chennai metropolitan area in recent years. Hence in the present study, the surface O<sub>3</sub> concentration was measured in this urban site, Chennai. The surface ozone was found to often reach higher levels in urban area of Chennai. This study was conducted at Koyembedu, which houses Chennai's mofussil bus terminus; hundreds of buses and other vehicles ply daily and hence vehicular emission is very high. This site is surrounded by a number of industrial areas located within a short radius. Further, a wastewater purifying plant is also located in close proximity.

According to the frequency of rainfall, a year can be divided into four seasons. The period between January and February represents the winter months and the period between March and May summer months. Southwest monsoon (SWM) is between June and September, and October to December is the period of northeast monsoon (NEM). Figure 2 *c* shows that the seasonal variation of O<sub>3</sub> at this site. While the lowest seasonal average was observed during winter (12.2 ppbv), the highest average was during summer (16.3 ppbv). During the SWM season the average was 15.4 ppbv followed by NEM season with 14 ppbv. Low concentrations observed during the winter season as a result of higher NO<sub>x</sub> scavenging, lower photochemical reactions due to lower intensity of solar radiation and lower ambient temperature lead to a decrease in the concentration of O<sub>3</sub>. The destruction of O<sub>3</sub> is increased in NEM was attributed to the non-availability of adequate solar radiation due to cloudy skies, and also the reduction in precursors species from the atmosphere by rain which took place during this season<sup>5</sup>.

During the study period, the average maximum day temperature was about 34.1°C observed in summer and average minimum temperature of about 27.38°C was observed during monsoon. The humidity reached its highest daily average value of 82% and the lowest daily average was 37.5%. Further, the hourly average of wind speed varied from a minimum of 2.3 m/s to a maximum of about 10.2 m/s during the study period. The daily average was in the 0.24–5.5 m/s range.

## Methods

Surface ozone measurements were carried out daily and ten measurements were made on all days between 08.00 h and 17.00 h (IST) during the period from June 2011 to September 2012. Furthermore, wind speed, temperature, relative humidity and UV-radiation were also measured simultaneously. Rainfall and cloud cover data were obtained from Regional Meteorological Centre, Nungambakkam, Chennai.

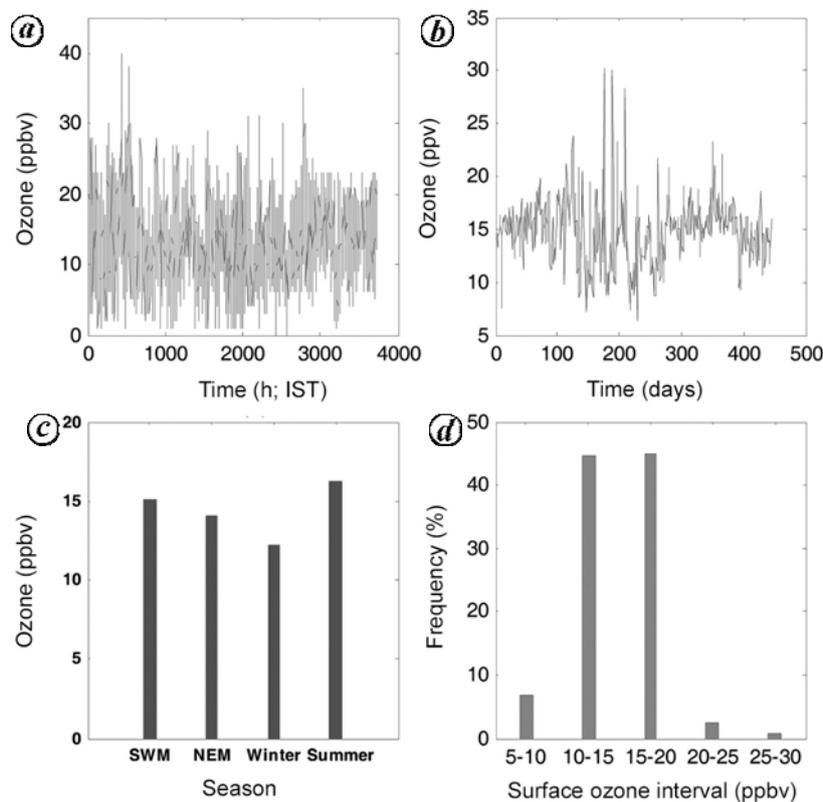
An aeroqual series 200 ozone monitor was used to measure low and high surface ozone levels. Its low concentration ozone head measures the ozone concentration

from 0.000 to 0.500 ppm and a high concentration ozone head measures ozone concentration from 0.050 to 20.00 ppm. Accuracy of a low concentration ozone head is  $\pm 0.010$  ppm (from 0 to 0.100 ppm)  $\pm 10\%$  (from 0.100 to 0.500 ppm), while that of a high concentration ozone head is  $\pm 10\%$  (from 0.20 to 2.00 ppm),  $\pm 15\%$  (from 2.00 to 20.00 ppm), the measurement units being either ppm or mg/m<sup>3</sup>. The sensor type is a gas sensitive semiconductor (GGS), which works on the principle of absorption of UV-radiation by ozone in the ambient air. The ozone sensor was calibrated against a certified UV photometer. The particular instrument was chosen for its simplicity and reliability in operation and for its ease of handling, cost-effectiveness and speed in obtaining gas concentration level directly. This has been of assistance in estimating the concentration of ground-level ozone in places where there was no permanent measurement. An aeroqual monitor with GSS ozone sensor has been used by several workers for the measurement of atmospheric ozone and nitrogen dioxide<sup>24–27</sup>. UV-radiation had been measured by UV light meter (UV-3450A series). Both instruments were supplied by Unipro Instruments India Pvt Ltd, Mumbai. Digital Anemometer Ms.6250Ms was used for wind speed measurement. Temperature and relative humidity were measured using thermometer and humidity meter.

## Results and discussion

The ozone data and all other corresponding parameters were analysed on the basis of daily and seasonal variations. For the discussion of diurnal variation, all 24 h readings were measured on an hourly basis on the 5th and 6th of every month. We have been measuring hourly instantaneous surface ozone since June 2011 to date and have used it for analysing data from June 2011 to September 2012. Night-time measurements had been made on some days of the month and it was found that surface ozone levels were very low during the night (<10 ppbv). The hourly values and daily average values were used to analyse the day-to-day variability. Monthly mean was calculated to study the seasonal cycle (Figure 2).

The overall distribution of datapoints O<sub>3</sub> is given in Figure 2 *a* with respect to hourly timescale. From the graph it can be seen that the accumulation of datapoints is high in the 10–20 ppbv range and varied from minimum 5 ppbv to maximum 40 ppbv. From the time series graph (Figure 2 *b*) of ozone concentration with respect to daily timescale, it is seen that accumulation of data is high in the 15–18 ppb range. It is clear that daytime average (08.00–17.00 h) of ozone concentration varied gradually from 7 to 30 ppbv, and the highest peak 30 ppbv was attained in the summer season. It can be seen from Figure 2 *d* that the highest surface ozone distribution was attained at 11–15 and 16–20 ppbv (40% each) and the lowest ozone distribution at 26–30 ppbv (2%).



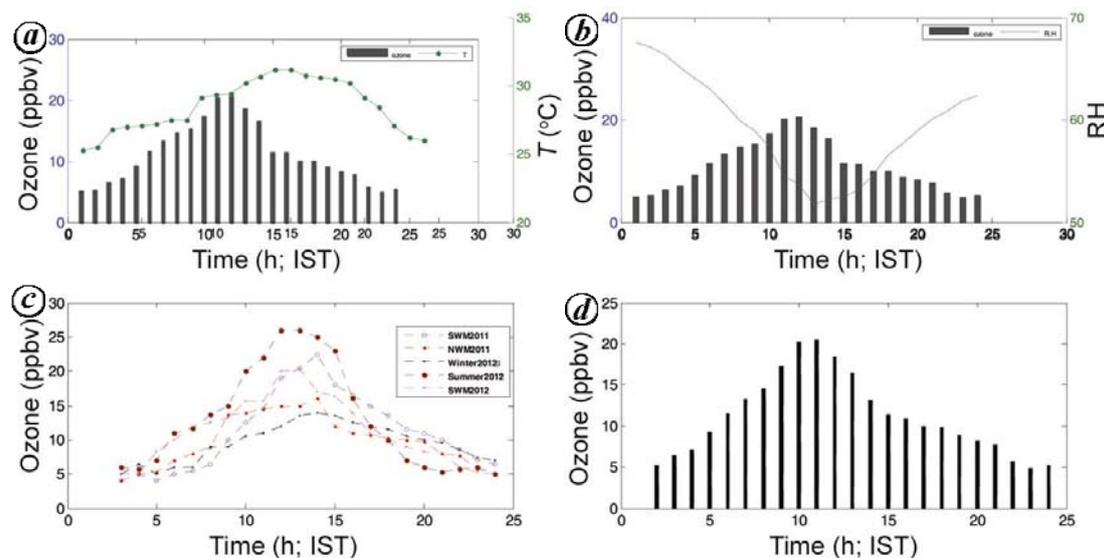
**Figure 2.** *a*, Hourly variation of surface ozone during the study period containing 3720 datapoints. *b*, Daily variation of surface ozone. *c*, Seasonal average of surface ozone. *d*, Frequency distribution of surface ozone during the entire study period.

### Diurnal and seasonal variation of ozone

From Figure 3, it can be observed that surface ozone production during night is quite low. The ozone concentration begins to increase after sunrise around 09:00 h (IST) and attains its highest level (25 ppbv) in the afternoon around 13:00–15:00 h mainly due to the photolysis of  $\text{NO}_2$  (ref. 8). During late evening hours, i.e. around 18:00–24:00 h the surface ozone concentration tends to decrease and reaches its lowest value of about 5 ppbv. Low value of  $\text{O}_3$  during night could be mostly due to the absence of photo-dissociation and its destruction through titration with NO and surface deposition<sup>1</sup>. The low concentrations of ozone observed during morning hours are due to the lower boundary layer height which mainly reduces the mixing process between the ozone-poor surface layer and the ozone-rich upper layer<sup>12</sup>. The rate of production of surface ozone during winter months is found to be minimum in an urban location. This can be attributed to the shrinking of the boundary layer height, as well as relatively low precursor gas concentration at the surface layer. The net loss of ozone observed rather late in the evening is due to slow titration of ozone because of low  $\text{NO}_x$  concentration. Further, a considerable amount of vehicular emission at this site makes it a rich  $\text{NO}_x$  environment where the titration process of ozone would domi-

nate more than its production<sup>12</sup>. Concentration of  $\text{O}_3$  increases with high temperature and low relative humidity (Figure 3 *a* and *b*). High temperature increases the emission of  $\text{NO}_x$  from soil<sup>3</sup>, which may be one of the reasons for increase in  $\text{O}_3$  concentration with temperature.

The variation in ozone concentration in different seasons may be due to the variation in  $\text{NO}_x$ , CO,  $\text{CH}_4$ , hydrocarbon levels and changing meteorological conditions like temperature, cloud cover, wind velocity, wind direction, relative humidity and rainfall<sup>5</sup>. The seasonal diurnal pattern has been determined by taking the overall average of the diurnal ozone values of various months. It could be observed that the pattern is almost similar during all the seasons, with a rise in ozone mixing ratio after sunrise and further attaining highest value during noon. However, it can be clearly observed that there is a significant difference in the average ozone values for various seasons. During summer, the diurnal pattern showed a peak followed by SWM and almost similar values during monsoon and winter periods. Such behaviour could be expected due to its direct relationship to the availability of precursor gases and favourable meteorological conditions. It can be seen that in summer the highest mean ozone concentration was 25 ppbv and lowest was 5.7 ppbv. For SWM the highest value was 22.5 ppbv and the lowest was 6.7 ppbv. The maximum and minimum



**Figure 3 a–d.** Diurnal variation of (a) ozone and temperature ( $T$ ) and (b) ozone and relative humidity (RH). c, Seasonal average of diurnal cycle of surface ozone. d, Diurnal cycle of surface ozone alone.

mean ozone concentration in winter season was 14 and 6.4 ppbv respectively. Moreover, NEM values were lower than that of SWM at 15 ppbv and 5 ppbv respectively.

### Variation of ozone with meteorological parameters

The variation in surface ozone concentration depends on not only on precursor emissions but also on meteorological conditions. Meteorological variables such as near surface wind, temperature and precipitation influence the ozone formation, deposition and transport process by affecting photochemical reactions and atmospheric dynamic conditions<sup>18</sup>. The influence of available meteorological variables on the surface ozone concentration at the observational site is discussed briefly in the following sections (Table 1, Figure 4).

### Correlation between temperature and surface ozone

The scatterplot in Figure 4a shows the relation between temperature and ozone in different seasons. A positive correlation is found in almost all the seasons. The  $r$  (correlation coefficient) values and  $m$  (slope) values are given in Table 1. Ambient air temperature differs with seasons of the year and time of the day. The photochemical production of surface ozone is a direct consequence of the ambient temperature and solar flux, and its concentration increases with respect to both parameters<sup>5</sup>. During the study period, the average maximum day temperature was about 34.1°C which was observed in summer and average minimum temperature of about 27.38°C was

observed in monsoon. In all the seasons it was observed that the highest temperature was reached at 15 : 00 h.

### Correlation between relative humidity and surface ozone

Figures 3b and 4b and Table 1 clearly show the relation between relative humidity and ozone in different seasons. When the humidity is higher, the major photochemical pathways for removal of ozone will be enhanced. From Table 1, it is clear that the relative humidity shows negative correlation with surface ozone in all the seasons. Furthermore, correlation between ozone and humidity is more pronounced in monsoon than summer. Hence the ozone concentration has a strong dependence on humidity. The humidity reaches its highest daily average value of 82% in monsoon season and the lowest daily average value of 37.5% in summer season.

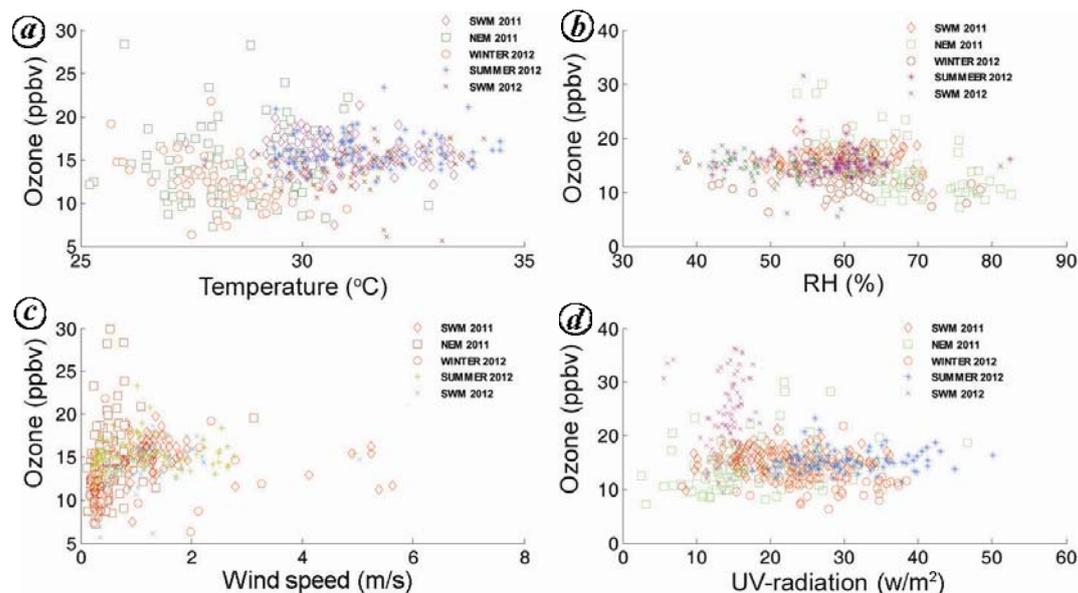
### Correlation between wind speed and surface ozone

It is known that there is a clear relationship between ambient air quality and wind speed. This parameter is important for the dispersion and transport of ozone and its precursors from their emission sources. At the place of study hourly average wind speed varied from a minimum 2.3 m/s to a maximum of about 10.2 m/s during the study period. The daily average was in the 0.24–5.5 m/s range. August and September experienced maximum wind speed. From Table 1 and Figure 4c it is clear that in the study area wind speed had a positive correlation with surface ozone concentration during most of the seasons. The

**Table 1.** Correlation between ozone and meteorological parameters

Season	Correlation between ozone and temperature		Correlation between ozone and wind speed		Correlation between ozone and relative humidity		Correlation between ozone and UV-radiation	
	<i>r</i>	<i>m</i>	<i>r</i>	<i>m</i>	<i>r</i>	<i>m</i>	<i>r</i>	<i>m</i>
SWM 2011	0.032	0.05	-0.191	-0.3	-0.05	-0.023	0.05	0.01
NEM 2011	0.057	0.19	0.27	0.75	-0.56	-0.37	0.25	0.15
Winter 2012	0.063	0.04	0.14	0.62	-0.28	-0.1	-0.063	-0.03
Summer 2012	0.132	0.18	0.22	0.44	-0.024	-0.083	0.11	0.05
SWM 2012	0.22	0.41	0.34	1.2	-0.62	-0.11	0.064	0.02

*r*, Correlation co-efficient; *m*, slope value; SWM, Southwest monsoon; NEM, Northeast monsoon.



**Figure 4 a-d.** Relation between ozone and temperature (a), RH (b), wind speed (c) and ozone and UV-radiation (d) in different seasons.

reason for a negative correlation in SWM can be attributed to insufficient wind dynamics and local wind circulation patterns that could not make any alteration on the dispersion and transportation of surface ozone<sup>3</sup>.

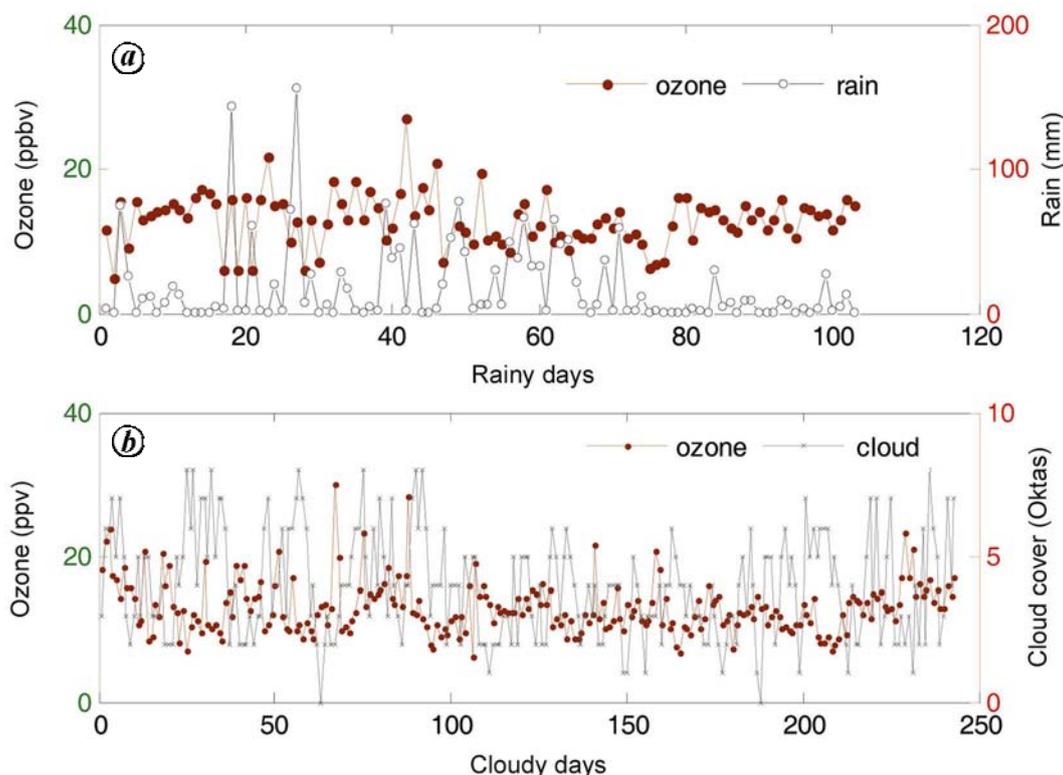
### Correlation between UV-radiation and surface ozone

The solar radiation that reaches the Earth's surface consists of wavelengths between 295 and 3000 nm. Sunlight is commonly separated into three main wavelength ranges; ultraviolet, visible and infrared. Wavelengths between 295 and 400 nm are considered the UV portion of the solar spectrum and it is between 4% and 7% of the total radiation. The spectral range of UV and sub-components are: UV-A (315–400 nm), UV-B (280–315 nm) and UV-C (<280 nm). Ozone is a secondary pollutant and it is not usually emanated in a straightforward manner from the stacks. Under suitable ambient meteorological conditions (e.g. warm, sunny, clear day), UV-radiation causes the precursors to interact photochemically in a set of reactions that results in the formation of ozone (eqs (5) and (6)).

Table 1 and Figure 4 d show the correlation between UV-radiation and ozone in different seasons of the year 2011 and 2012. During all seasons, except winter the ozone concentration increases with UV-radiation. In winter, it shows a negative correlation. This is because in winter, the absence or presence of sunlight being unimportant, due to higher atmospheric instability the photochemical process slows down and surface ozone gets depleted. In the study area, day average maximum UV radiation of 42 w/m<sup>2</sup> was experienced during summer season and day average minimum of 3.5 W/m<sup>2</sup> was experienced during NEM.

### Correlation between rainfall and surface ozone

The most prominent meteorological feature in the study area was monsoon rainfall. The SWM which usually sets in by the first week of June lasts till September. This is followed by NEM which lasts till December. On the average about 90% of the total rainfall occurs from June to December, which constitutes the monsoon season. The months of January and February with insignificant rain



**Figure 5.** Relation between day average of surface ozone and day average of (a) rainfall and (b) cloud cover.

and moderate relative humidity represent winter season. While the period between March and May represents summer in which high convective activities are observed due to high temperature (Figure 5).

Figure 5a shows the influence of daily average rainfall on daily average ozone. The concentration of surface ozone attained high value when the rainfall was low and it attained comparatively lower value when maximum rainfall occurred. Correlation coefficient has been found in rainy days only. The overall correlation between average rainfall and average ozone is  $-0.14$ . Concentration of  $O_3$  is affected significantly by rain. Ozone levels tend to be higher under hot and sunny conditions which are favourable for photochemical ozone production. Conversely, wet rainy weather with higher relative humidity is typically associated with low ozone levels provided by wet ozone deposition on the water droplets.

### Correlation between cloud cover and surface ozone

Clouds can bring about changes in solar radiation that are responsible for increasing or decreasing photochemical reaction in the troposphere and thus reduce or enhance the troposphere ozone concentration. They can directly absorb ozone and its precursors in cloud liquid water, thereby decreasing the tropospheric ozone<sup>2</sup>. Figure 5b shows that the concentration of surface ozone attained higher value during low cloud occurrence and compar-

tively lower value at both higher and moderate cloud occurrence. It is found that the overall correlation between cloud occurrence and ozone concentration ( $r$ ) is  $-0.159$ . This indicates that cloud occurrences which affect the amount of solar radiation reaching the surface of the Earth are important for photochemical reactions in the formation of ozone in the study area.

Cloud occurrence shows the number of days and/or nights, the sky is fully or partially covered with clouds during various times in each month. Cloud occurrence attained lowest monthly average of 2.7 Oktas in March; then gradually increased and attained maximum monthly average of 5 Oktas in October and gradually decreased.

### Conclusion

During the period of study we were able to collect data containing 3720 datapoints of hourly reading and 466 daily reading at Chennai, a tropical urban site, in the southeastern coastal region of India. The data have been analysed from which we can infer the following.

Diurnal variation of ozone and variation of ozone along with rainfall, cloud cover, temperature, wind speed and relative humidity in different months have been studied. Therefore changes in these parameters due to climate change will necessarily impact surface ozone concentration. To assess the relation with meteorological parameters, the correlation coefficients between the surface-ozone concentration and meteorological variables were

calculated. The diurnal pattern of surface ozone explains that ozone production depends on the photochemical production process.

It was observed that the hourly average value of surface ozone attained maximum (40 ppbv) during hot summer months. Also, it attained maximum daily average value of 30 ppbv during the summer months. But, a previous study<sup>3</sup> had shown that maximum hourly average 53 ppbv was attained in May 2005. Even though there is no indication of an increasing trend in surface ozone concentration and the ozone level never exceeded the national ambient air quality standard of 75 ppbv during the present study, there is a strong probability that it will reach unsafe levels in future. Hence we should minimize the sources of ozone precursors.

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