Variability of surface ozone with cloud coverage over Kolkata, India

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Critical analysis of experimental surface ozone data and cloud coverage is reported over Kolkata during the period January 2011 to December 2011. Significant relationship between these two parameters is observed. Analysis shows that the trend of surface ozone concentration and cloud coverage follow opposite tendency. Some exceptional observations are also reported. Apart from this, seasonal variation of surface ozone with columnar ozone concentration, NO₂ and temperature is also examined in relation with cloud cover to obtain a possible explanation of this type of variation.

1. Introduction

Ozone plays a very important role in tropospheric chemistry. Though present in trace amounts (~0.0012%) in the atmosphere (Iqbal 1983), it influences the weather and climate of a place, thus acting as an important greenhouse gas (Kiehl *et al.* 1999; Rex *et al.* 2004). Solar ultra-violet (UV) radiation has several harmful effects on human health (Kondratyev and Varotsos 2001). The wavelength covering UV radiation (200–400 nm) is subdivided into three spectral regions: UV-C (200–280 nm), UV-B (280–315 nm) and UV-A (315–400 nm) (Vanicek *et al.* 2000; World Health Organization 2002). Of these, UV-C is completely absorbed by the stratospheric ozone. UV-B is influenced by changes in the column ozone whereas UV-A is least absorbed. Thus ozone also plays a key role in protecting the living beings on the earth by absorbing the most energetic part of the solar ultraviolet radiation (Alexandris *et al.* 1999; Varotsos *et al.* 1995). The amount of UV-B and

UV-A consequently determines the quantity of ozone that will be formed in the troposphere. The important factors influencing the ground level UV radiation are clouds, solar zenith angle, total column ozone and surface albedo (Burrows 1997; Kerr and McElroy 1995). Research has shown that cloud coverage controls the short-term variability of the UV-radiation in the mid-latitudes (Calbó *et al.* 2005). Nevertheless, cloudiness frequently disguises the influence of ozone changes on UV radiation and may alter the expected UV signal (Glandorf *et al.* 2005), in turn affecting the ground level ozone concentration. Thus clouds act as one of the most significant factors influencing solar radiation, and consequently the photolysis rate constants. The water droplets or the ice-crystals in the clouds enhance the scattering of the radiation resulting in loss or in a reduced atmosphere transmissivity (López *et al.* 2009). Hence cloud levels may have a daily variability of the UV levels at the surface (Ziemke *et al.* 1997). Broken cloud situation may also arise when ground level

Keywords. Surface ozone; cloud coverage; NO₂; OMI.

Figure 1. Percentage frequency of occurrence of cloud coverage (%) over 24-hr of the day over Kolkata for 2011.

Figure 2. (a). Monthly variation of surface ozone with cloud coverage for January 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

radiation is found to be higher in cloudy conditions than in cloudless situations (Estupiñán *et al.* 1996). Due to its characteristic variability, it is difficult to predict the effects of clouds on the UV radiation. Consequently, the effect of cloud coverage is mostly taken as an approximate value and it is also location specific. In Kolkata, past studies have been conducted on the variation of total column ozone with temperature (Midya *et al.* 2011) and different seasons (Midya and Saha 2011). Effects of cloud coverage and total column ozone over different cities in India report that concentration of column ozone increased with the increase of cloud occurrence (Jana *et al.* 2010, 2011). The primary objective of this paper is to evaluate the effect of cloud coverage on the surface ozone over Kolkata.

2. Materials and methods

2.1 Site description

The experimental set-up is placed at Jadavpur University Salt Lake (JUSL) campus in the city of Kolkata (22°33'N, 88°30'E; 6 m amsl); the second largest metropolis in south Asia. The city has a population of 4.5 million (http://www.censusindia.gov.in/2011-prov-results/prov_data_products_wb.html). The campus is situated in the greater Kolkata region in the eastern part of the city (Ghosh *et al.* 2013). The surrounding locality has mainly residential buildings and offices. Dhapa, the municipal solid waste landfill is located ~2 km to the south of the site. The airport is also located ~10 km to the north-east of the study location.

Figure 2. (b). Monthly variation of surface ozone with cloud coverage for February 2011 at (i) 00Z, (ii) 03Z (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

Figure 2. (c). Monthly variation of surface ozone with cloud coverage for March 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

Several small scale industries are scattered in different parts of the city such as heavy engineering, mineral processing, pharmaceuticals, food processing, agriculture, electronics, textiles, etc. (Ghose *et al.* 2004). Kolkata also experiences substantial level of pollution from automobiles (Mallik *et al.* 2014). To the west of the city, lie two coal fired power plant stations of Budge Budge (~30 km from site) and Kolaghat (~70 km from site). Besides this, Haldia, the port city located about 100 km to the south-west of Kolkata has a major refinery and pharmaceutical company. Other industrial cities located in the vicinity of Kolkata are Durgapur and Asansol, which are situated about 170 and 210 km respectively to the north-west of Kolkata. They have major power plants and several industries. Thus local emissions from

automobiles and the local small scale industries along with the emissions of these distant industrial regions collectively account for the overall emission scenario of the city of Kolkata.

The city experiences a tropical wet and dry climate according to Köppen climate classification. Köppen classification is an extensively used empirical climate classification system based on vegetation zones. This classification subdivides the terrestrial climate into five major climate groups, namely A, B, C, D, and E*, each having various types and subtypes. All these types are governed by temperature criteria except B, which is defined by

*Gr A: Tropical/Megathermal; Gr B: Dry; Gr C: Temperate/Mesothermal; Gr D: Continental; Gr E: Polar and Alpine.

Figure 2. (d). Monthly variation of surface ozone with cloud coverage for April 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

dryness (<http://www.britannica.com/EBchecked/topic/322068/Koppen-climate-classification>). The tropical wet-dry climate (abbreviated as Aw), as experienced in Kolkata, is characterized by distinct wet and dry seasons. Temperatures are high throughout the year and also have a wide range. Precipitation occurs mostly in the summer season. Further details of the city's meteorology are already discussed in Ghosh *et al.* (2013) and Mallik *et al.* (2014).

2.2 Experimental set-up

The experimental set-up consists of an ambient ozone analyzer (Model 49i; Make: Thermo Fisher Scientific, USA) installed at the study site to carry

out online measurement of ambient ozone. A 4 m-long teflon pipe connects the analyzer to the outside air through a peltier unit. The peltier unit absorbs moisture and the resultant dry air is passed through a 5 μm pore size Polytetrafluoroethylene (PTFE) filter paper for dust filtration after which the air enters the sample inlet of the instrument. The ozone analyzer operates on the principle that ozone molecules absorb UV light at a wavelength of 254 nm. The degree to which the UV light is absorbed is directly related to the ozone concentration according to Beer–Lambert Law. The Model 49i calculates the ozone concentration and makes the data available as analog outputs. The lower detectable limit is 1 ppbv and the data are stored as a 15-min average.

The NO_2 data for a complete year (2011) is collected from WBPCB.

2.3 Satellite and meteorological data

Along with the surface trace gas data, columnar ozone data is also collected from the NASA Giovanni Portal (disc.sci.gsfc.nasa.gov/giovanni). Columnar ozone data over Kolkata were collected from Ozone Monitoring Instrument (OMI) onboard the Aura satellite. This study employs the seasonal study of columnar ozone taken from OMI Level 3 with a resolution of $1.0^\circ \times 1.0^\circ$ (latitude $\times$ longitude).

The cloud coverage data is collected from the NOAA Air Resources Laboratory (ARL) website (<http://ready.arl.noaa.gov/READYamet.php>).

Other meteorological data are also obtained from University of Wyoming website (<http://weather.uwyo.edu/>) for all necessary calculations.

2.4 Methodology

This paper incorporates the study of surface ozone and cloud coverage for one year – 2011. The NOAA ARL website gives the cloud coverage data as percentage (%) measured at every 3 hrs on a day starting from 00Z. This measurement only accounts for the area of the sky covered with clouds and does not consider other details such as cloud type, albedo or solar radiation receipt. Since we are considering surface ozone production, which is photochemistry driven, only data of 00Z (0530 Local

Figure 2. (e). Monthly variation of surface ozone with cloud coverage for May 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

Time – LT), 03Z (0830 LT), 06Z (1130 LT) and 09Z (1430 LT) are considered for computation here. The photochemical production of ozone is at its best during these times, hence the selection. Along with the surface ozone data, the columnar ozone values are also analysed to find any correlation. Besides this, the seasonal variation of ozone with primary precursor – NO_2 and temperature has also been observed.

3. Results and discussions

This study entitles continuous monitoring of surface ozone and cloud coverage over Kolkata during a

year long period from January to December, 2011. At first, the frequency distribution of cloud coverage is observed (figure 1). The distribution, out of 365 occurrences, mostly shows either clear sky conditions (0–10%) or fully covered sky condition (90–100%) during the entire study period. Thus, we may say that during instances of clear sky condition, presence of surface ozone concentrations is mainly governed by photochemical production induced by high intensity of solar radiation. In case of partially or fully covered sky, the substantial amounts of ozone present may be accounted for due to other meteorological effects. We shall discuss in detail these cases in the coming sections.

Figure 2. (f). Monthly variation of surface ozone with cloud coverage for June 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

Figure 2. (g). Monthly variation of surface ozone with cloud coverage for July 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

3.1 Monthly variation of ozone and cloud coverage

The monthly variations of surface ozone and cloud coverage for all the months of the study period are plotted and shown in figure 2(a–l). The x-axis represents the date of every month, the primary y-axis characterizes surface ozone values which are plotted as columns (blue) and the secondary y-axis represents the cloud coverage values (red dots). For each month, the variations of surface ozone with cloud coverage are plotted at 00 to 21Z. Among all the months studied, the best results are obtained from end of October to February. These months cover the post-monsoon and the winter period of this region (according to the guidelines of India

Meteorological Department). In winter, Kolkata experiences moderately warm days and cool nights as this region falls in the Tropical wet-dry region according to Koppen classification. The cloud coverage over eastern India is lesser than the other times of the year and for the maximum time it is cloudless. This is clear from the figures as in most of the cases cloud coverage values show very low percentage of coverage. Due to clear sky condition, maximum solar radiation reaches the earth surface around this time, so the surface ozone content increases. In most of the cases, it is observed that the concentration of surface ozone and cloud coverage is inversely related. The result is expected as surface ozone is photochemically produced and in

Figure 2. (h). Monthly variation of surface ozone with cloud coverage for August 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

the presence of clouds less amounts of UV radiation penetrates to reach the surface and hence the production is less.

Ozone near the surface is formed through a complex series of reactions involving the action of sunlight in presence of nitrogen oxides (NO_x) and volatile organic compounds (Haagen-Smit 1952; Crutzen 1979). Profuse amounts of CO, non-methane hydrocarbons (NMHCs), and NO_x are found in the tropical region due to large-scale biomass burning. Substantial levels of these gases are also released from automobile emissions.

Tropospheric ozone forms by reactions of oxides of nitrogen (NO_x) and volatile organic compounds

(VOCs) in the presence of solar radiation. Ozone photochemistry occurs when NO_2 is photolyzed in sunlight. Ozone formation is the result of the following reactions (Seinfeld and Pandis 2006):

Once formed, ozone reacts with NO to regenerate NO_2 .

Figure 2. (i). Monthly variation of surface ozone with cloud coverage for September 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

Net ozone production is not possible unless a peroxy radical is present to react with NO to regenerate the NO₂ without destroying an ozone molecule as in reaction (3).

(where R can be: H, CH₃, C₂H₅, etc.) (4)

In the Indian subcontinent, higher water content in the atmosphere, also favours the photochemical production of ozone. Besides transport and dispersion, air mass plays a pivotal role in formation and building up of ozone.

3.2 Seasonal variation of surface ozone with its precursor, columnar ozone and meteorological parameters

The seasonal variation of surface ozone, its precursor NO₂ and columnar ozone over Kolkata is presented in figure 3. The monthly average values of surface ozone (blue line in the plot) in 2011 shows a more or less smooth trend with higher values in the winter period and lower during the monsoon. The monthly average of its precursor – NO₂ (red line in the plot) – shows a maximum at the beginning of the year, during winter (January–February 2011). The values steadily decrease during the

Figure 2. (j). Monthly variation of surface ozone with cloud coverage for October 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

pre-monsoon months (March–May 2011) and attain a minimum during the monsoon period. From late monsoon onwards (September–October 2011), surface NO_2 concentration again starts building up. The trend of the columnar ozone concentration as obtained from the OMI shows maximum values during the pre-monsoon months (March–May 2011). If we minutely observe the trends of both the surface and the columnar ozone values, we find a similarity in the changes of concentration during March–June 2011, when both show a gradual decreasing tendency. Again during September–October 2011, the concentration increases and then decreases till December 2011. Thus it may be suggested that even though in a

very small extent, the columnar ozone concentration of Kolkata has a certain amount of influence on the overall surface concentration of ozone. The seasonal variation among the surface ozone and NO_2 concentration, however, show no direct correlation amongst themselves.

Along with the seasonal variation of the gases, seasonal temperature profile and CC values are also computed and plotted in figure 4. The blue and the red lines represent the monthly average temperature and CC over Kolkata for the entire year of 2011. As expected, the CC is maximum during the monsoon months: June–September 2011. The highest amount of clear sky is observed during the winter, followed by post-monsoon. The average

Figure 2. (k) Monthly variation of surface ozone with cloud coverage for November 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

monthly temperature illustrates the typical profile that of a region of tropical wet and dry climate. The lowest temperatures are recorded during the winter months followed by the post-monsoon. For rest of the year, the temperature remains more or less high in the range of 25°–35°C.

3.3 Correlation study

Correlations between surface ozone and cloud coverage are also studied and presented in figure 5. Since there is a time lag between the amount of ozone formed to the cloud coverage, the surface ozone data of 30 min later than the cloud

coverage are considered for computation. The x-axis and y-axis represent CC (%) and surface ozone (ppb), respectively. The coefficient of determination (R^2) of ozone *vs.* cloud coverage shows very poor correlation. The R^2 values at 00Z, 03Z, 06Z, 09Z, 12Z, 15Z, 18Z and 21Z are 0.032, 0.0, 0.105, 0.159, 0.018, 0.061, 0.063, and 0.042, respectively. An interesting trend is obtained. From 06Z to 12Z, there is a negative correlation among surface ozone and CC. This indicates that during daytime (06Z–12Z), amount of ozone at the surface is driven by photochemical production from its precursors. Hence, higher the CC, lesser is the amount of surface ozone. Again as night approaches (15–00Z), the trend is reversed. This signifies that

Figure 2. (I). Monthly variation of surface ozone with cloud coverage for December 2011 at (i) 00Z, (ii) 03Z, (iii) 06Z, (iv) 09Z, (v) 12Z, (vi) 15Z, (vii) 18Z and (viii) 21Z.

Figure 3. Seasonal variation of surface ozone, NO₂ and columnar ozone over Kolkata for 2011.

Figure 4. Seasonal variation of temperature (T) and cloud coverage (CC) over Kolkata for 2011.

in the absence of sunlight, other meteorological parameters have an active role to play in the presence of surface ozone concentration. Although very poorly correlated, the significance of such poor correlations among these two parameters may be that surface ozone production is not primarily dependant on the cloud coverage at our site. Other meteorological parameters along with different emission sources are responsible. These results are also supported by the fact that during the study periods several anomaly days have been observed on which the trend between surface ozone and cloud coverage have been found just the reverse. More about the anomaly days is discussed in the following section.

Figure 5. Correlation between surface ozone (ppb) and cloud coverage (%) at (a) 00Z, (b) 03Z, (c) 06Z, (d) 09Z, (e) 12Z, (f) 15Z, (g) 18Z and (h) 21Z.

Table 1(a). *Basis of assumption for the selection of anomaly days.*

	Threshold Values		
	03Z	06Z	09Z
	Ozone (ppb) [Median – 15.37]	Ozone (ppb) [Median – 27.37]	Ozone (ppb) [Median – 29.43]
Anomaly due to high ozone and high CC (>50%)	>23.05	>41.05	>44.15
Anomaly due to low ozone and low CC (<50%)	<7.68	<13.69	<14.72

Table 1(b). *Dates on which anomaly were observed due to high ozone and high cloud coverage.*

Date	CC (%)	Ozone (ppb)
03Z		
16.10.11	52.9	34.25
26.09.11	53.4	36.54
08.05.11	55.9	23.53
01.02.11	59.1	26.10
21.10.11	59.1	31.28
25.09.11	60.4	36.28
28.05.11	62.0	23.57
05.09.11	62.1	24.48
18.10.11	63.5	35.84
01.09.11	74.6	26.59
19.10.11	77.5	30.01
03.01.11	78.9	24.27
30.07.11	82.5	23.31
23.07.11	82.9	25.05
27.09.11	84.4	39.96
08.09.11	84.8	24.61
15.08.11	90.8	24.02
02.08.11	92.3	26.63
24.07.11	94.8	23.35
16.07.11	95.2	23.24
19.08.11	96.2	24.31
18.08.11	96.3	24.41
14.07.11	97.8	23.43
29.07.11	97.9	23.47
15.07.11	99.2	23.88
16.08.11	99.4	24.77
11.08.11	99.5	23.13
10.06.11	99.6	24.69
06Z		
21.10.11	51.2	57.21
25.09.11	51.9	61.96
26.09.11	56.0	51.04
01.02.11	58.8	53.88
23.03.11	62.7	42.90
18.10.11	71.5	61.49
24.09.11	73.6	47.33
27.09.11	76.6	46.89
19.10.11	77.3	52.24
15.08.11	88.0	55.28
01.08.11	94.0	43.23
10.06.11	96.6	43.63
09.06.11	97.3	50.04

Table 1(b). *(Continued.)*

Date	CC (%)	Ozone (ppb)
09Z		
29.09.11	53.2	45.85
16.10.11	54.3	47.62
12.01.11	55.1	44.27
17.10.11	59.8	51.36
28.01.11	60.6	61.00
21.10.11	63.6	64.69
27.09.11	70.0	45.44
30.11.11	78.0	44.86
27.11.11	81.3	45.13
26.09.11	83.1	49.49
23.09.11	88.4	45.43
18.10.11	96.1	56.04
10.06.11	96.5	64.34
19.10.11	96.9	55.42
24.09.11	98.0	57.89
01.12.11	99.5	50.43
09.06.11	100.0	56.40

3.4 Anomaly observed in the correlation study

Regardless of the good results, certain anomaly days have been observed which are tabulated in table 1. The table represents the anomaly dates at three different times – 03Z, 06Z, and 21Z. The assumptions based on which the anomaly days selected are tabulated in table 1(a). The anomalies may be categorized basically into two types: when ozone concentrations are higher despite high CC (table 1b) and low ozone concentrations with low CC (table 1c). In both the cases, the time period of 00Z and 12–21Z are neglected. This is due to the fact that since we are focussing on ozone production, the phenomenon of photochemical production of ozone is not very predominant at these hours in this part of the world. Thus the amount of surface ozone present at these times is mainly contributed from either residual amounts from previous days production or from advection of air mass from nearby or distant areas.

Table 1(c). *Dates on which anomaly were observed due to low ozone and low cloud coverage.*

Date	CC (%)	Ozone (ppb)
03Z		
13.02.11	0	3.56
22.02.11	0	7.61
25.02.11	0	4.45
13.03.11	0	3.03
11.04.11	0	6.44
14.11.11	0	5.20
17.11.11	0	4.66
23.11.11	0	6.14
11.12.11	0	7.35
12.12.11	0	5.94
15.11.11	0.3	2.51
10.12.11	1.3	2.22
15.03.11	3.6	6.61
01.01.11	4.9	4.74
10.03.11	5.7	2.67
14.01.11	8.2	6.10
14.02.11	14.3	4.88
30.01.11	15.6	4.81
02.11.11	18.3	1.14
05.03.11	18.8	6.10
16.11.11	25.1	3.26
27.03.11	25.2	5.56
04.12.11	27.1	1.61
17.02.11	31.2	6.62
02.12.11	33.6	4.39
07.01.11	34.2	7.59
11.03.11	34.2	6.67
28.11.11	36.8	6.95
01.11.11	39.4	1.13
03.12.11	46.6	2.56
29.03.11	47.3	5.14
06Z		
14.03.11	0	9.86
15.11.11	0.2	13.28
10.03.11	5.3	10.88
14.01.11	7.7	9.44
06.11.11	10.3	12.48
14.12.11	12.6	13.43
27.03.11	18.1	13.25
02.11.11	18.4	3.48
13.12.11	26.1	10.16
04.11.11	26.7	10.25
21.12.11	35.0	12.68
01.11.11	35.5	2.88
09Z		
14.11.11	4.6	13.34
04.11.11	11.6	10.84
14.12.11	14.4	13.51
01.11.11	23.4	2.84
02.11.11	32.0	9.05
13.12.11	39.3	13.72
13.10.11	41.7	12.85
22.12.11	42.2	12.87
11.09.11	47.3	7.92

These anomaly situations can only be justified on the basis of horizontal wind transport. Though insolation is the driving force for ozone production in the troposphere, concentrations of the precursors are also a matter of concern here. In case of low ozone concentrations with low cloud coverage, it may be suggested that even with high insolation, either the precursor concentrations were low or the ozone produced was dispersed due to horizontal wind transport. On the otherhand, in the opposite case, high ozone values may result from either windborne transport or due to certain meteorological conditions, which have prevented the previously produced ozone from getting dispersed.

4. Concluding remarks

Clouds are an essential component of weather on Earth. They are condensed atmospheric moisture the form of minute water droplets or ice crystals. In addition to solar UV radiation and precursor gases, cloud coverage plays a major role in contributing to the ground level ozone of a place. Under no cloud or high transparent cloud cover condition, more insolation reaches the ground level favouring the photochemical processes that generate ground level ozone. So, the expected results should be with increase in cloud cover, ground level ozone decreases. The presence of UV radiation is an essential condition for the production of surface ozone by NO₂ photolysis reaction. The high cloud coverage prevents the UV radiation to reach the surface and for this reason less amount of surface ozone is formed. The study over Kolkata shows good connection between surface ozone and cloud coverage. Apart from this relation, the seasonal variation of surface ozone along with its precursor NO₂, columnar ozone, and temperature indicate a highly complex relationship among these species. Even though, good relation was obtained at times, certain anomalies were observed, which clearly indicate the dominant effect of other meteorological parameters responsible for these anomalies. Thus all these parameters are singularly ineffective in explaining the presence of surface ozone over Kolkata. Rather, a combined effect of all of them along with other meteorological aspects such as convective transport, inversion, etc., is helpful in elucidating the anomaly instances.

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