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Research Paper

Effect of Cloud occurrences on Tropospheric and Stratospheric Ozone over Alipore (22.52°N, 88.33°E) and Bangalore (12.97°N, 77.56°E), India

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Abstract: *The paper presents the nature of annual cycles of tropospheric and stratospheric ozone, and cloud occurrences, monthly variations of tropospheric and stratospheric ozone, and cloud occurrences, and variations of tropospheric and stratospheric ozone with occurrences of cloud for the period January, 2005 to December, 2011 Alipore (22.52°N, 88.33°E) and Bangalore (12.97°N, 77.56°E), India. Annual cycle of low- level cloud occurrences depicts that the low-level cloud over Alipore and Bangalore had been noticed to occur for many days and nights, particularly in the months from June to September. The low level cloud occurrences were found in winter months and post-monsoon period. The concentration of tropospheric ozone over Alipore had a greater value than that over Bangalore, but the concentration of stratospheric ozone over Alipore was relatively less. The effect of cloud occurrences on tropospheric and stratospheric ozone concentration over Alipore and Bangalore has been critically analyzed, explained and compared. It has been observed that the concentration of tropospheric ozone had a decreasing tendency with cloud occurrences, whereas the stratospheric ozone has a slight increasing trend with the increase of cloud occurrences and vice versa. The related possible chemical and physical explanation of role of cloud occurrences on tropospheric and stratospheric ozone has been offered.*

Keywords: Tropospheric and stratospheric ozone, cloud occurrences.

Introduction:

Ozone though a minor, but one of the dominant constituent in the atmosphere controls atmospheric environmental quality and atmospheric chemical processes. The concentration of ozone in the stratosphere is gradually declining, whereas the tropospheric counterpart shows a tendency to increase persistently due to human activities. Stratospheric ozone plays an important role in protecting mankind and the environment by absorbing harmful solar UV-B radiation partially and UV-C radiation totally, but in the troposphere a too high amount of this species has a detrimental effect on human being and also to the environment.

Tropospheric ozone, a key chemical constituent serves as a product and the participant of the photochemical reactions therein. The variation in concentration and distribution in ozone exerts an immediate effect on life time and distribution of other atmospheric chemical species such as SO_2, NO_2 and OH radicals, thereby influencing the composition and equilibrium of tropospheric chemistry.

Clouds are also a vital element of weather on Earth. Without cloud weather would not exist. Today, there are different forms or genera of clouds recognized by the International Cloud Classification, and there are three height categories with an established altitude range for each category. Low-level clouds range from the surface to 6,500 ft (2,000 m), mid-level from 6,500-23,000 ft (2,000-7,000 m), and high-level generally above 20,000 ft (6,000 m). High-level clouds are Cirrus (Ci), Cirrus uncinus, Cirrus Kelvin-Helmholtz · Cirrostratus (Cs), Cirrocumulus (Cc), Cirrocumulus undulatus, Pileus and Contrail. Mid-level clouds are Altostratus (As), Altostratus undulatus, Altocumulus (Ac), Altocumulus undulatus, Altocumulus mackerel sky, Altocumulus castellanus and Altocumulus

lenticularis. Low-level clouds consist of Fog, Stratus (St), Cumulus (Cu), Cumulus humilis (Cu), Cumulus mediocris (Cu), Stratocumulus (Sc), Arcus (Roll), Fractus, Funnel, Nimbostratus (Ns), Nimbostratus virga, Shelf, Wall, Actinoform cloud, Undulatus asperatus and vertical clouds are Cumulonimbus (Cb), Cumulonimbus incus, Cumulonimbus calvus, Cumulonimbus mammatus, Cumulus congestus, Cumulus castellanus, Pyrocumulus, Pyrocumulonimbus, Overshooting top and Accessory.

The clouds are condensed atmospheric moisture in the form of minute water droplets or ice crystals. Condensed nuclei can be anything varying from dust to debris. Pratt [1] detected some mineral dusts and primary biological particles, such as bacteria, pollen and fungi, that can behave as ice nuclei and initiate the formation of ice-crystals in clouds. A large group of condensation nuclei makes the cloud visible. The cloud comprises water droplets. The liquid water content of the cloud can be defined as the mass of water in the condensed state per unit volume. It is also referred [2] as the total water content and not only the water in droplet form. Several researchers [3-6] have reported that liquid water content in cloud varies from 0.10g/m^3 to 1 g/m^3. Carrier [7] and Slobin [3] have reported that water droplet concentration varies from 70 per cm^3 to 465 per cm^3. It has been observed by Slobin [3] that the average diameter of the water droplet in cloud varies from 9 µm to 20 µm while Carrier [7] reported that the drop size alters from 1 µm to 60 µm. The cloud contains saturated water vapour. The cloud occurs at higher altitude also. The cloud has liquid water content between 0.1 and 1.0 g/ m3. The thickness of the cloud is usually between 1.5 km and 2.5 km, depending on the type of the cloud. Slobin [3] reported that for the light cloud the thickness is around 0.2 km while for medium cloud the thickness is 0.5 km and for heavy cloud the thickness is 1 km and 1.5-2.0 km. The cloud occurrence morphology over Bangalore and Dumdum has been obtained from low level cloud of about 2-6 km altitude.

The formation and destruction of ozone in the troposphere comprises a series of complex cycles in which atomic oxygen, molecular oxygen, carbon monoxide, oxides of nitrogen, water vapour, volatile organic compounds etc. are involved. Ozone in the troposphere is produced by the addition of ground state oxygen atoms O (^{3}p) to molecular oxygen assisted by any third body M to ensure simultaneous momentum and energy conservation [8].

$$O\left(^3p\right) + O_2 + M \rightarrow O_3 + M$$

The primitive terrestrial atmosphere was oxygenic in nature and ozone was produced by photodissociation of water vapour as follows [9]:

$$H_2O + h\nu \rightarrow OH + H$$
$$OH + h\nu \rightarrow O + H$$
$$O + O + M \rightarrow O_2 + M$$
$$O + O_2 + M \rightarrow O_3 + M$$

The source of oxygen atoms in the troposphere is not same as in the stratosphere where oxygen atoms are made by the photo dissociation of O_2 at UV wavelengths less than 240nm. In the troposphere, only UV radiation with greater than 290nm is available, because of essentially complete absorption of shorter wavelengths of O_2 and O_3 above the tropopause. Atomic oxygen in the troposphere is produced by photo dissociation of NO_2 and tropospheric O_3 as follows:

$$i) NO_2 + h\nu(\lambda = 424nm) \rightarrow NO + O$$
$$ii) O_3 + h\nu(\lambda = 300 - 320nm) \rightarrow O + O_2$$

Tropospheric ozone is mainly formed by photochemical oxidation of carbon monoxide, methane and non-methane volatile organic compounds (NMVOCs) in the presence of nitrogen oxide radicals (NOx ≡ NO + NO_2). Oxidation begins with the reaction of CO with OH radical. The hydrogen atom thus produced rapidly reacts with oxygen molecules to give peroxy radicals. Peroxy radicals then continue to react with NO to NO_2 that is photolysed to atomic oxygen which reacts with O_2 to make a molecule of O_3.

[O_2]

$$OH + CO \rightarrow H + CO_2$$
$$H + O_2 \rightarrow HO_2$$
$$HO_2 + NO \rightarrow OH + NO_2$$
$$NO_2 + h\nu(\lambda = 424nm) \rightarrow NO + O$$
$$O(3P) + O_2 + M \rightarrow O_3 + M$$

The net effect of these reactions is:

$$CO + 2O_2 \rightarrow CO_2 + O_3$$

The OH radical in the troposphere is produced by photo dissociation of nitrous, nitric acids, ozone [10] and hydrogen peroxide [10] as follows:

$$HONO + h\nu(\lambda = 400nm) \rightarrow OH + NO$$
$$HNO_3 + h\nu(\lambda = 350nm) \rightarrow OH + NO_2$$
$$O_3 + h\nu(\lambda \leq 320 - 410nm) \rightarrow O(1D) + O_2$$
$$O(1D) + H_2O \rightarrow 2OH$$
$$H_2O_2 + h\nu \rightarrow 2OH$$

Peroxy radical is also made from the photolysis of formaldehyde [10].

$$CH_2O + 2O_2 + h\nu \rightarrow CO + 2HO_2$$

SO_2 injected into the atmosphere may also ozone formation by absorbing radiation. SO_2 absorbs radiation strongly between 180 nm and 235 nm, weakly between 260-340 nm and very weakly between 340 to 390 nm [11].

$$SO_2 + h\nu \rightarrow SO + O(\lambda \leq 220nm)$$
$$SO + O_2 \rightarrow SO_2 + O$$
$$2(O + O_2 + M \rightarrow O_3 + M)$$

Net: $3O_2 \rightarrow 2O_3$

The simplified form of troposoheric ozone formation from volatile organic compounds (VOCs) obeys the following steps:

$$VOC + OH + O_2 \rightarrow RO_2 + H_2O$$
$$RO_2 + NO + O_2 \rightarrow NO_2 + HO_2 + CARB(\text{sec}ondaryVOC)$$
$$HO_2 + NO \rightarrow NO_2 + OH$$
$$2(NO_2 + O_2 + h\nu \rightarrow NO + O_3)$$

Net:
$$(NO_x + OH) + VOC + 4O_2 \rightarrow 2O_3 + CARB + H_2O + (NO_x + OH)$$

Where, R denotes organic radicals and CARB stands for carbonyl compounds.
The loss of tropospheric ozone is mainly due to the following reaction [10]:

$$O_3 + h\nu(\lambda \leq 320 - 410nm) \rightarrow O(1D) + O_2$$

OHx radicals are removed from the atmosphere by the formation of hydrogen peroxides, organic peroxides, nitric acids and other organic nitrates.

$$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$$
$$RO_2 + HO_2 \rightarrow RCOOH + O_2$$
$$OH + NO_2 \rightarrow HNO_3$$

Stevenson [12] proposed the higher ozone production rates in the troposphere due to higher NOx emission, higher isoprene emission, more detailed NMHC scheme and improved parameterization of process such as photolysis, convection and stratosphere-troposphere exchange. From the above mechanisms of tropospheric ozone production and loss, it is clear that solar radiation plays a key role. The main factors that affect the atmospheric radiative transfer at wavelengths important for tropospheric ozone budget are the solar zenith angle, absorption by stratospheric ozone, reflection from the surface, scattering and absorption by aerosol particles and clouds. From the observation of the short wave radiation effects of clouds on the tropospheric ozone, Voulgarakis [13] found that clouds have a modest effect on ozone on global scale, but their role is much more significant on a regional scale. They observed that both the production and loss of ozone increase significantly above the cloudiest areas and decrease below. The main cause of this feature is that the photolysis rates of NO_2 and O (^{1}D) decrease below the clouds because of attenuation of radiations by cloud particles and increase above due to backscattering. Solar zenith angles are small in the tropics and in such cases the increases in photolysis rates start at the lowest parts of the clouds.

The cloud chemistry is composed of gaseous phase chemistry, aqueous phase chemistry and scavenging of soluble gases [14]. The clouds can bring about the changes in solar radiation that is responsible for increasing or decreasing photochemical reaction in the troposphere and thus reduce or enhance the tropospheric ozone concentrations. It can directly absorb ozone and its precursors (NOx, NMHC, free radicals etc.) in cloud liquid water thereby declining tropospheric ozone [14]. It provides various aqueous phase chemical reactions happening to species absorbed by cloud, responsible for the changes in gaseous phase ozone concentration. Clouds provide surfaces for heterogeneous chemistry to take place [15]. Precipitating clouds scavenge soluble trace gases and aerosols from troposphere [14]. The vertical motions associated with clouds result in substantial convective transport of chemical species. Particularly, deep convection can provide an important source of hydrogen oxide radicals in the upper atmosphere, leading to enhanced production of ozone [16]. Lightning associated with the deep convective clouds is an important source of nitrogen oxides in the middle and upper atmosphere [17].

Cloud can also scatter and absorb incoming solar radiation, modifying the actinic flux and thus photolysis frequencies of key chemical species. The enhanced photolysis frequencies have been observed above and in the upper level of clouds, while reduced frequencies were found below optically thick clouds and absorbing aerosols [18]. Cloud reduction of solar radiation is wavelength dependent in the UV-range. The longer wavelength UV-A is more strongly attenuated than UV-B wavelengths [19]. Reduced photolysis frequencies occur underneath a cloud. The UV radiation reflected up from the cloud can enhance the upwelling actinic flux by 50% to 150% which results 30-40% increase in the total above-cloud photolysis frequencies [20]. In addition, short-term local enhancement of photolysis frequencies of about 40% has been observed at the Earth's surface under some specific broken cloud conditions where the disc of the sun is not occluded by a cloud and the cloudy portion of the sky is brighter than the clear sky portion [21].

Clouds in the troposphere can influence the photolysis rates (*J* values) and hence concentrations of chemical species. Tie *et al* [10] suggest that global mean OH concentration increases by about 20% due to the impact of clouds. Because of clouds, the globally averaged photolysis rates of *J* [O_3], *J* [CH_2O} and *J* [NO_2] are enhanced in the troposphere by about 12, 13, and 13%, respectively, that lead to an 8% increase in the tropospoheric ozone. *Liu et al* [22] proposed that the global mean change in OH concentration is insignificant (~1%), but it shows much larger changes above (5-10%) and below (-5-20%) the

tropical deep convective clouds and the mid-latitude low-level clouds, as well as near surface (approximately -20%). For O_3, the global mean effect is about 3-5% increase. O_3 increase in the tropical upper troposphere is ~5-8%. They suggested that O_3 increase above the clouds is due to backscattering of solar radiation, whereas O_3 increase below the tropical deep clouds is due to the reduced net O_3 losses. Voulgarakis *et al* [13] reported that the largest averaged change in chemical budgets of ozone was found in marine troposphere where cloud optical depths were high. Cloud effects were small on average in the middle troposphere because this is a transition region between reduction and enhancement in photolysis rates. Increases in boundary layer ozone due to clouds were driven by large scale changes in downward ozone transport from higher in the troposphere rather than by decreases in in-situ chemical loss rates. Increases in upper tropospheric ozone are caused by higher production rates due to back scattering of radiations and consequent increases in photolysis rates of mainly NO_2.

It is now well established that different environmental parameters may influence the variation of ozone concentration in different layers in the atmosphere. Ozone layer in the atmosphere also plays an important role in controlling different events occurring in troposphere, stratosphere and mesosphere. Ozone is not equally distributed in the atmosphere. The concentration of ozone gradually increases from upper troposphere of about 10 km altitude, attains maximum value at an altitude of about 25 km and then gradually decreases. Dobson *et al* [23] observed that the average temperature of the earth is directly related to ozone concentration of stratosphere and fall in ozone concentration in the stratosphere takes place over England before the arrival of warm front at the ground surface. The rise in ozone content in the stratosphere occurs when cold front reaches near the ground level. Mitra [24] reported the close relation between barometric height, tropospheric weather and ionospheric parameters of the upper atmosphere. It was observed that the minimum height of F region and average E ionization tend to follow the variation of barometric height. Correlation was observed between the lowest virtual height of E region and ground temperature at Standford, California, USA. Bates [25] and Mackay *et al* [26] reported that the variation of solar UV radiation due to fall of stratospheric ozone concentration can influence tropospheric climate in several ways. Midya and Sarkar [27] observed the correlation of the variation of stratospheric ozone with relative humidity and sharp depletion of absolute humidity related with Nor'wester over Kolkata. Mooley and Parthasarathy [28] analyzed all-India summer monsoon (June to September) rainfall for the period 1871 to 1978. They reported that the highest and lowest rainfall country level was observed in the years 1961 and 1877, respectively. There was a continuous rise in 10-yr mean rainfall from 1899 to 1953. Rakecha and Soman [29] observed that the annual extreme rainfall records of most of the stations over India were free from trend and persistence. The extreme rainfall series at stations over the west coast, north of 12^0 N and some stations to the east of the Western Ghats over the central parts of the Peninsula showed a significant increasing trend at 95% level of confidence. Stations over Southern Peninsula and over the lower Ganga valley had been found to exhibit at decreasing trend at same level of significance.

Monod and Carlier [30] performed a box model study of the multiphase photochemistry (both gas and aqueous phases) of C_1 organic compounds within a non-precipitating cloud on a local scale. They found that in a situation where ozone accumulates in clear sky conditions, as soon as cloud is formed, the tropospheric ozone estimate changes drastically, the net production decreases by a factor of 2 or more and depending on the NOx concentration and the pH values, can actually lead to chemical destruction. Bremaud and Taupin [31] investigated the influence of clouds on the diurnal cycle of ozone concentration in the Reunion Island marine boundary layer. They reported that the orographic cloud layer plays an important role in the chemistry of ozone through several ways. The two major ones are the enhancement of photodissociation processes in the interstitial air relative to clear sky conditions and the perturbation of the NOx photostationary state through the scavenging of HO_2 and CH_3O_2 radicals. They concluded that the greater destruction of the marine boundary layer ozone observed in Reunion Island relative to the mean 4 ppbv was due to clouds. Williams and Toumi [32] reported that the inclusion of high cloud was found to bring about warming of the troposphere, resulting in a net heating in the lower stratosphere. It strengthened the circulation, leading to decrease in total tropical ozone. Jana *et al* [33] reported the significant rise of total atmospheric ozone column density with increase in cloud occurrences not considering other meteorological parameters. Jana et al [34] also presented the nature of annual cycles of tropospheric ozone, cloud occurrences, NO_2, rainfall, SO_2, SPM, CO, NMHC (non-methane hydrocarbon) and surface solar radiation for the period October' 2004 to June' 2009 and explained over Alipore (22.52^oN, 88.33^oE), India.

The effect of these micro-meteorological parameters including cloud occurrences on tropospheric ozone had been critically analyzed. Possible explanation based on physical considerations and chemical kinetics had been critically discussed. In this paper, the variations of monthly mean tropospheric and stratospheric ozone, and cloud occurrences, annual cycles of these climatic parameters, and the effect of cloud occurrences on tropospheric and stratospheric ozone from January 2005 to December 2011 over Alipore and Bangalore has been shown and compared and explained. Alipore, an important site in the south of the mega city Kolkata is situated on the east bank of the river Hooghly. The Sea, Bay of Bengal is about 50 km away from it in the south. The population is approximately 4600000 over an area of 185 sq. km around it. The city is significantly polluted over a large number small scale industry, vehicular traffic, thermal power plants and the very busy ports. At the west bank opposite to

Kolkata, there is Howrah city which is full of small and large industries of iron and jute mill. Bangalore (12.97°N, 77.56°E), the heart of the Mysore Plateau (a region of the larger Precambrian Deccan Plateau) at an average elevation of 900 m (2,953 ft), lies in the southeast of the South Indian state of Karnataka. It covers an area of 741 km^2. The topology of Bangalore is generally flat, though the western parts of the city are hilly. Bangalore has a tropical savanna climate with distinct wet and dry seasons. Due to its high elevation, Bangalore usually enjoys a more moderate climate throughout the year, although occasional heat waves can make summer somewhat uncomfortable.

Data and analysis: TOMS (total ozone mapping spectrometer) version 8 monthly mean column density of tropospheric ozone in Dobson unit (DU) is derived from the Convective Cloud Differential (CCD) Method [35] over Alipore station in India. Tropospheric column ozone can usually be determined from following two well known approaches from satellite data. In the first method, stratospheric column ozone is derived by combining Upper Atmosphere Research Satellite (UARS) halogen occultation experiment (HALOE) and microwave limb sounder (MLS) ozone measurements. Tropospheric column ozone is then obtained by subtracting these stratospheric amounts from the total column. Total column ozone in this study includes retrievals from Nimbus 7 (November 1978 to May 1993) and Earth probe (July 1996 to present) total ozone mapping spectrometer (TOMS). Data from HALOE are used in this first method to extend the vertical span of MLS (highest pressure level 46 hPa) using simple regression. This assimilation enables high resolution daily maps of tropospheric and stratospheric ozone which is not possible from solar occultation measurements alone, such as from HALOE or Stratospheric Aerosols and Gas Experiment (SAGE). Ziemke *et al* [35] provided another new and promising technique that yields tropospheric column ozone directly from TOMS high density footprint measurements in regions of high convective clouds. They defined this method as the convective cloud differential (CCD) technique. The CCD method uses TOMS total ozone measurements over highly reflecting, high altitude clouds. In some regions, especially the tropical western Pacific, these high-reflectivity clouds are often associated with strong convection and cloud tops near the tropopause. The tropospheric column can be obtained at cloud-free pixels by subtracting the above-cloud stratospheric ozone amount from TOMS total ozone. Because cloud height information is not measured by TOMS, the primary assumption in the CCD method is that the high-reflectivity clouds often have cloud tops at the tropopause. The monthly mean ozone concentration in DU (Dobson Unit) at Alipore and Bangalore has been obtained from the website http://jwocky.gsfc.nasa.gov, published by NASA, USA for the period January' 2005 to December' 2011.

The cloud data over Alipore (22.52°N, 88.33°E) and Bangalore have been collected from the Indian Meteorological Department, Kolkata and Pune, India and belongs to the period January' 2005 to December' 2011. The cloud cover is estimated by the observer and expressed on a scale [36] ranging from 0 to 8 Octas. Hence 0 Octas means clear sky, 4 Octas means that one - half (or four-eighths) of the sky is covered with cloud and 8 Octas implies fully covered (overcast).

Results and Discussion

Annual cycles of monthly mean tropospheric and stratospheric ozone concentrations and the number of days and / or nights, the sky is fully or partially covered with cloud during various time over Alipore (22.52°N, 88.33°E) and Bangalore, India for the period January 2005 to December 2011 have been presented by Figure 1.

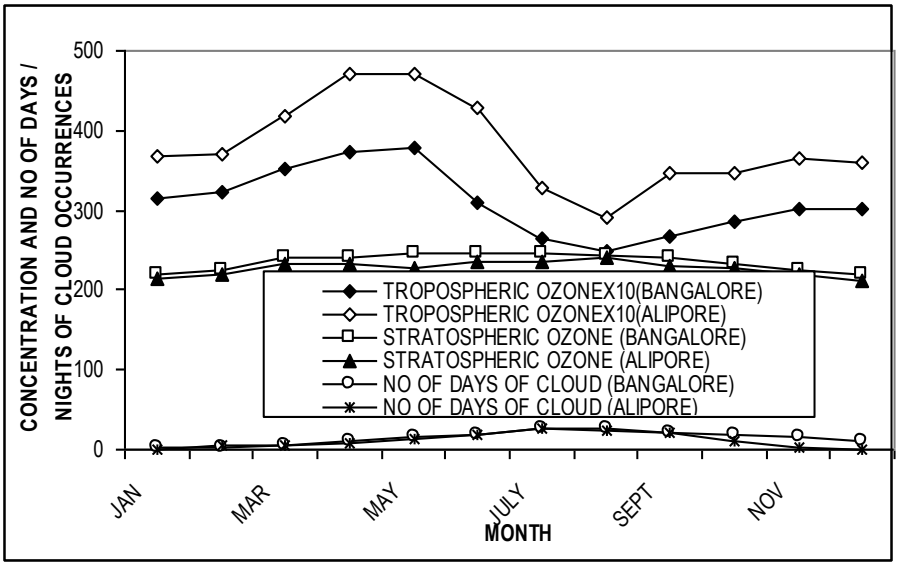

Figure 1: Annual cycles of tropospheric and stratospheric ozone, number of days/nights of cloud occurrences over Alipore (22.52°N, 88.33°E) and Bangalore (12.97°N, 77.56°E) for the period January, 2005 to December, 2011

The nature of annual cycles of monthly mean tropospheric ozone followed the same trend over Alipore and Bangalore (Corr. coeff 0.93), but the concentration of tropospheric ozone over Alipore was higher than that over Bangalore. Amount of tropospheric ozone concentration gradually increased from the month of January, attained maximum for the months of May, then gradually decreased, achieved minimum value for the month of August and then increased very slightly. This is due to the fact that the rates of the reactions which favor the tropospheric ozone formation from its precursors were enhanced by sharp increase of surface solar radiation from January to May. During August, the minimum amount of tropospheric ozone was as a result of lower photolysis rates of ozone formation processes caused by lower surface radiation energy due to large occurrences of clouds in the monsoon time (JJAS) as well as lower concentration of ozone precursor viz. CO, NMHC, SO_2, NO_2 etc. After the month of August, the rate of increase in concentration of ozone was slow because of disappearance of cloud, mild fall of solar radiation and steady rise of ozone precursor amount. The relatively lower tropospheric ozone over Bangalore had been attributed to relatively lower surface radiation energy, larger occurrence of cloud, lower concentration of ozone precursors, higher altitude and lower rate of ozone formation processes. Annual cycles of stratospheric ozone over Alipore and Bangalore followed identical trend for the same period (Corr, coeff 0.93). The concentration of ozone had increased gradually from the month of January, attained the maximum amount during the months of May and June and then gradually declined. This trend was due to gradual increase of solar radiation from January to May and June that favored the formation processes of ozone, and gradual decline of solar radiation which favored the ozone depletion processes in the stratosphere. The concentration of stratospheric ozone over Bangalore was slightly higher than that over Alipore because of greater rate of ozone formation and lower rate of ozone depletion over Bangalore.

Cloud occurrences show the number of days and / or nights, the sky is fully or partially covered with cloud during various time in each month. Nature of annual cycle of cloud occurrences over the above two stations obeyed nearly the identical trend (Corr. coeff. 0.89). Cloud occurrences attained the lowest value in the month of January, then gradually increased from the month of January, attained its highest value in the months of July and August, then gradually decreased. Amount of rainfall increased exponentially from January to July, attained the maximum value in the month of July and then gradually decreased. Comparatively high rainfall was observed from June to September. In case of the station Alipore, low occurrences of cloud and rainfall before and during pre-monsoon (MAM) had been attributed to occasional existences of nor'wester that occurred usually 2-6 times in each month from February to May in every year. Large cloud occurrences and rainfall during monsoon (JJAS) were obviously due to the existences of monsoon wind from middle of June that carried huge amount water vapour from south oceans consisting of Bay of Bengal, Indian Ocean and Arabian Sea. Gradual fall of cloud occurrence and rainfall during post-monsoon was as a result of dry-retreat monsoon or north- westerly wind comprising very less amount of water vapour entering from Bihar, Orissa, Assam including Bangladesh that might clash with wet wind of Bay of Bengal causing cyclone sometimes due to the high pressure and temperature difference over Alipore. In case of the station Bangalore, comparatively more occurrence of cloud at pre-monsoon, monsoon and post-monsoon season was due to frequent thunderstorm during pre-monsoon (March-May) and southwest and north east monsoon wind loaded with huge amount of water vapour from Bay of Bengal, Indian Ocean and Arabian Sea during monsoon (June-September) and retreat monsoon wind during post-monsoon (October-November) carrying larger amount of water vapour from Bay of Bengal.

Solar radiation exponentially increased from January to May, achieved maximum in the month of May and then slowly decreased. The reason behind an enhancement in surface solar radiation from January to May is the gradual decrease in distance between the earth and the sun and increase in daytime from 21 st December to 21 st June. Sudden fall in surface solar radiation in the month of June was because of large occurrence of cloud appeared by the monsoon. Comparatively gradual lowering in solar radiation in monsoon and post-monsoon time was also due to large occurrence of cloud in monsoon, slow increase in distance between the Earth, decrease in daytime and the sun and appearance of fog and smog during Autumn and Winter time.

Variation of monthly mean tropospheric and stratospheric ozone, and cloud occurrences over Alipore and Bangalore from January, 2005 to December, 2011 has been depicted in Figure 2.

It reveals that variations of concentrations of tropospheric (Corr. Coeff. 0.77) and stratospheric ozone (Corr. Coeff. 0.9) over Alipore and Bangalore were oscillatory. Tropospheric ozone over Alipore was relatively higher, whereas stratospheric ozone was comparatively lower over Alipore for the same period. The concentration of tropospheric ozone at both these stations had risen very slowly for this period probably due to slow increase in the amount of ozone precursors and surface temperature because of enhanced surface solar radiation, but the rate of increase of tropospheric ozone was higher over Alipore due to greater amount of ozone precursors and relatively higher surface temperature that enhances ozone formation processes below the cloud. Again, the rate of increase in stratospheric ozone over Bangalore became relatively higher due to larger occurrences of cloud over Bangalore that caused greater backscattering of solar radiation promoting ozone formation processes in the stratosphere.

The variations of occurrences of clouds over Alipore and Bangalore were also oscillatory, but the cloud occurrences over Alipore had a decreasing tendency, whereas it had an increasing tendency over Bangalore from January 2005. Relatively higher tropospheric ozone and lower stratospheric ozone over Alipore was due to higher solar surface radiation, larger amount of tropospheric ozone precursors and lesser occurrences of clouds over Alipore. Scattered variations of tropospheric and stratospheric ozone over Alipore and Bangalore for the period January, 2005 to December, 2011 with cloud occurrences have been represented in Figure 3.

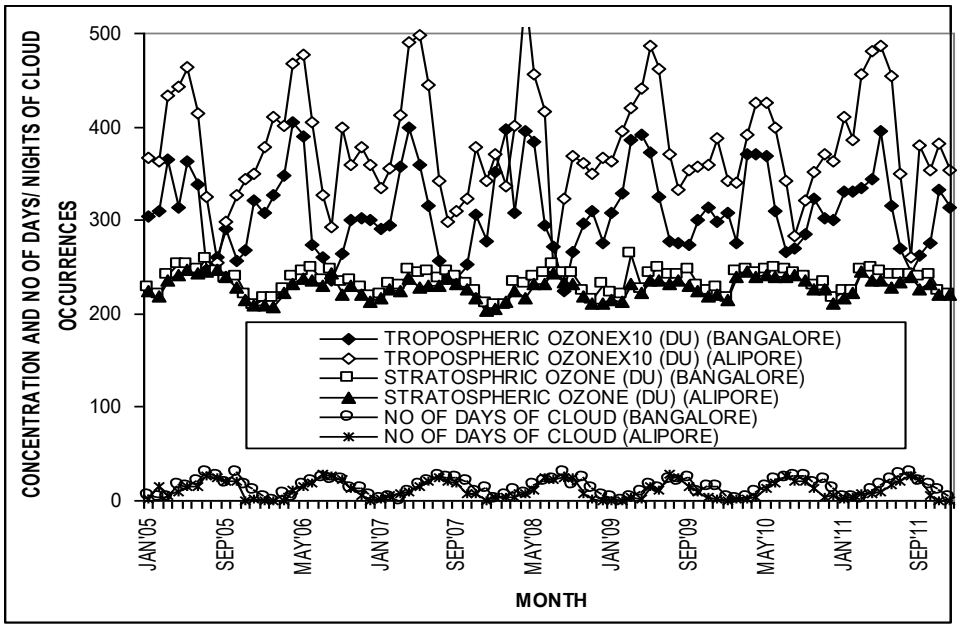

Figure 2: Variations of monthly mean concentrations of tropospheric and stratospheric ozone, number of days/nights of cloud occurrences over Alipore (22.52°N, 88.33°E) and Bangalore (12.97°N, 77.56°E) from January, 2005 to December, 2011

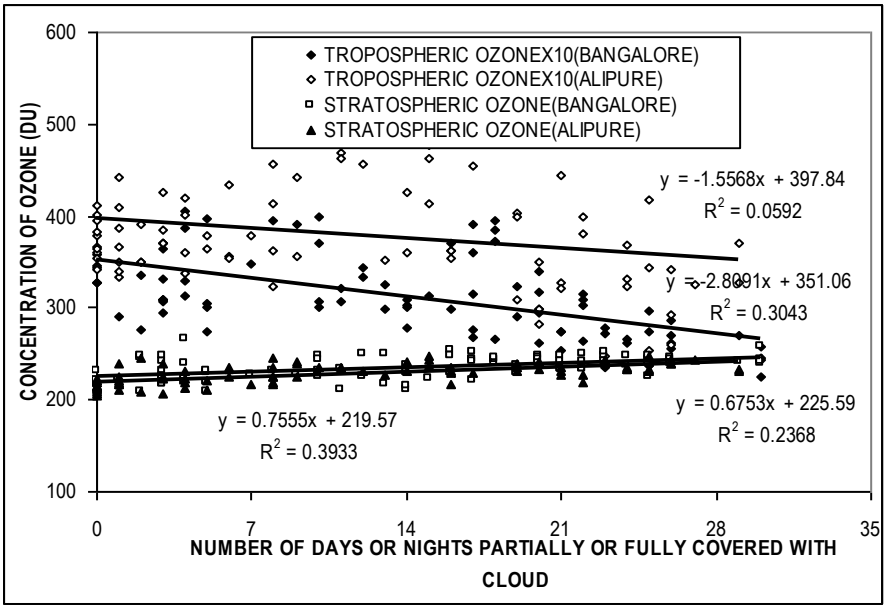

Figure 3. Scatterred diagram of concentrations of of tropospheric and stratospheric ozone with number of days/nights of cloud occurrences over Alipore (22.52°N, 88.33°E) and Bangalore (12.97°N, 77.56°E) for the period January, 2005 to December, 2011

It clearly indicates that tropospheric ozone concentration had decreased with the increase of cloud occurrences over

both these stations because of more absorption of solar radiation by larger clouds in the troposphere. But, the rate in fall of tropospheric ozone concentration with cloud was relatively higher over Bangalore. The stratospheric ozone concentration had increased with the increase of cloud occurrences at both these stations due to more backscattering of solar radiation above the cloud. But, the rate in increase of stratospheric ozone concentration with cloud was relatively lower over Bangalore.

Conclusion

Clouds can not only influence the earth's climate through modulation of the earth's energy and hydrological cycles but also tropospheric and stratospheric photochemistry through modification of solar radiation that determines photolysis rates for ozone production and loss. Annual cycles of tropospheric ozone and cloud occurrences over Alipore and Bangalore from January' 2005 to December' 2011 revealed a gradual rise in tropospheric ozone from January to May because of increasing rates of ozone formation reactions by the enhanced surface solar radiation, the minimum amount of tropospheric ozone in the month of August due to lower photolysis rates of ozone formation processes caused by lower surface radiation energy due to large occurrences of clouds in the monsoon time (JJAS) as well as lower concentration of ozone precursor viz. CO, NMHC, SO_2, NO_2 etc. and then slow increase in concentration of ozone due to disappearance of cloud, mild fall of solar radiation and steady rise of ozone precursors. The comparatively lower tropospheric ozone over Bangalore was due to the relatively lower surface radiation energy, larger occurrence of cloud, lower concentration of ozone precursors, higher altitude and lower rate of ozone formation processes. The concentration of stratospheric ozone over Alipore and Bangalore had increased gradually from the month of January, attained the maximum amount during the months of May and June and then gradually declined due to gradual increase of solar radiation from January to May and June that favored the formation processes of ozone, and gradual decline of solar radiation which favored the ozone depletion processes in the stratosphere. The concentration of stratospheric ozone over Bangalore was slightly more than that over Alipore because of greater rate of ozone formation and lower rate of ozone depletion over Bangalore.

Low occurrences of cloud and rainfall before and during pre-monsoon (MAM) over Alipore had been observed for the occasional existences of nor'wester usually occurred 2-6 times in each month from February to May in every year, large cloud occurrences and rainfall during monsoon (JJAS) obviously as a result of the existences of monsoon wind from middle of June that brought huge amount water vapour from south oceans and gradual fall of cloud occurrence and rainfall during post-monsoon due to dry-retreat monsoon or north- westerly wind comprising very less amount of water vapor entering from Bihar, Orissa, Assam including Bangladesh that might clash with wet wind of Bay of Bengal causing cyclone sometimes because of the high pressure and temperature difference. Comparatively more occurrence of cloud over Bangalore at pre-monsoon, monsoon and post-monsoon season was due to frequent thunderstorm during pre-monsoon (March-May) and southwest and north east monsoon wind loaded with more huge amount of water vapor from Bay of Bengal, Indian Ocean and Arabian Sea during monsoon (June-September) and retreat monsoon wind during post-monsoon (October-November) carrying larger amount of water vapor from Bay of Bengal.

Slow rise of monthly mean tropospheric ozone over Alipore from October, 2004 to June, 2009 was probably due to slow increase in the amount of ozone precursors and surface temperature because of enhanced surface solar radiation as a result of declining cloud occurrences.

Variation of monthly mean tropospheric and stratospheric ozone, and cloud occurrences over Alipore and Bangalore from January, 2005 to December, 2011 depicts that variations of concentrations of tropospheric and stratospheric ozone over Alipore and Bangalore though oscillatory had a slow increasing trend for this period probably due to slow increase in the amount of ozone precursors and surface temperature because of enhanced surface solar radiation, with the greater rate of increase of tropospheric ozone over Alipore due to greater amount of ozone precursors and relatively higher surface temperature that enhances ozone formation processes below the cloud. Again, the rate of increase in stratospheric ozone over Bangalore became relatively higher due to larger occurrences of cloud over Bangalore that caused greater backscattering of solar radiation promoting ozone formation processes in the stratosphere. The variations of occurrences of clouds over Alipore and Bangalore though oscillatory had a decreasing tendency over Alipore and an increasing tendency over Bangalore from January 2005.

Scattered variations of tropospheric and stratospheric ozone over Alipore and Bangalore for the period January, 2005 to December, 2011 with cloud occurrences clearly reveal that tropospheric ozone concentration had decreased with the increase of cloud occurrences over both these stations because of more absorption of solar radiation by larger clouds in the troposphere with relatively higher rate in fall of tropospheric ozone concentration with cloud over Bangalore. The stratospheric ozone concentration had increased with the increase of cloud occurrences at both these stations due to more backscattering of solar radiation above the cloud.

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