

Variability in Sun photometer–derived total ozone over a tropical urban station

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[1] A portable, handheld, multichannel Sun photometer (ozonometer) has been used to measure total column ozone over Pune ($18^{\circ}32'N$, $73^{\circ}51'E$, 559 m above mean sea level), India, a tropical urban station. Data collected on about 575 mostly clear-sky days during the 5 year period from May 1998 to May 2003 have been used in this study to examine diurnal and seasonal variations. The overall daily mean total ozone at this station is 254 Dobson units (DU), with a variability of about 7%, while the most frequently occurring value is in the range 241–250 DU. The diurnal (daytime) patterns are observed to be different in the winter (December–February), premonsoon (March–May), and postmonsoon (October–November) seasons. There exists a predominant seasonal oscillation in total ozone, with higher values (265 DU) during premonsoon months and lower values (246 DU) during winter months. Ground-based Sun photometer–derived total ozone variations compare very well with those obtained by Earth Probe–Total Ozone Mapping Spectrometer. **INDEX TERMS:** 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0394 Atmospheric Composition and Structure: Instruments and techniques; 1610 Global Change: Atmosphere (0315, 0325); 3374 Meteorology and Atmospheric Dynamics: Tropical meteorology; **KEYWORDS:** total column ozone, Sun photometer, tropical station

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1. Introduction

[2] Concentrations of ozone in the atmosphere can change as a result of both natural processes and human activities. Changes in stratospheric ozone are related to changes in tropospheric source gases, to variations in solar radiation, volcanic events, and to changes in large-scale transport. Changes in tropospheric ozone are mainly associated with increase in urban pollution and biomass burning. Stratospheric ozone has decreased over the last several decades, while tropospheric ozone has increased over the Northern Hemisphere midlatitudes [Logan *et al.*, 1999].

[3] About 40% of the global ozone is located in the tropics between $25^{\circ}N$ and $25^{\circ}S$, and a strong decadal variation of total ozone in the tropics with maxima approximately in phase with the 11 year solar cycle has been documented [World Meteorological Organization (WMO), 2003]. Also statistically significant negative trends in total ozone were found at all latitudes except possibly in the tropics, where ground-based measurements show no long-term trends [Madronich *et al.*, 1994]. From total column ozone data (Dobson spectrophotometer) for the period

1957–1994, Chakrabarty *et al.* [1998] have found no long-term trend over Pune. However, the Dobson ozone data of the same station for the period 1981–1998 showed a decreasing trend [Londhe *et al.*, 2003].

[4] There are numerous ways to measure ozone in the atmosphere, but they fall broadly into two categories: measurements of total or column ozone and measurements of the vertical profile of ozone. Regular measurements of total column ozone are available from a network of surface stations, mostly in the mid latitude Northern Hemisphere, with reasonable coverage extending back to the 1960s. Near global, continuous total ozone data are available from satellite measurements since 1979. The ratio of the intensity of direct sunlight at two wavelengths in the 300–320 nm range is a measure of the total abundance of ozone in a column through the atmosphere. This forms the basic operating principle for a variety of optical instruments that monitor atmospheric ozone. The two well-known ground-based ozone-monitoring instruments are the Dobson and Brewer spectrophotometers. They are universally accepted for measuring column ozone, but they are expensive, heavy and large in dimension. There has been a need for a more portable, compact instrument that can measure ozone with reasonable accuracy. In the present study a handheld, advanced filter ozonometer has been used by the authors to

measure total ozone and its variability on daily, monthly and seasonal scales at a tropical urban location.

2. Instrument and Methodology

[5] Optical ozone measuring instruments follow, in principle, the filter photometric technique. Ground-based ozone monitors (ozonometer) employing narrow-band-pass interference filters were first developed nearly 30 years ago [Oshervich *et al.*, 1969; Mathews *et al.*, 1974; Steblova, 1975]. However, the drawbacks in the early interference filter-based instruments were their wide bandwidth, temperature dependencies and aging of the filters, which produce a gradual decrease in percentage transmission and a shift in the center wavelength. Such filter degradation can occur due to excessive exposure to direct sunlight, known as solarization and also due to reaction with absorbed moisture. High-quality interference filters were used in the handheld filter ozonometer known as Total Ozone Portable Spectrometer (TOPS) by Mims [1992] which was later significantly modified in the late 90s into an advanced version known as MICROTOS-II [Morys *et al.*, 2001; Ichoku *et al.*, 2002; Knobelspiesse *et al.*, 2003]. This handheld, compact filter ozonometer (Microtops II, version 2.42) has been in operation at Pune (18°32'N, 73°51'E, 559 m AMSL), India, a tropical urban station since May 1998 for measurements of total ozone (in Dobson Units) besides simultaneously monitoring precipitable water and aerosol optical depth.

[6] The UV wavelengths employed for ozone measurement are 305.5, 312.5, and 320.0 nm having a FWHM band pass of 2 ± 0.3 nm. The ozonometer has the above three UV channels and two near-IR channels (940 and 1020 nm) for precipitable water measurements and aerosol optical depth at 1020 nm. A quartz window provides access to the collimator tubes for the five photodiodes and a Sun alignment target. A spring-loaded door protects the window when the instrument is not in operation. Initially the date, universal time, and geographical coordinates of the location must be entered manually into the instrument's keypad or automatically by a Global Positioning System (GPS) receiver. The observer opens the window door and points the ozonometer toward the Sun until a bright spot of light is centered over the cross hair arrangement in a Sun target window provided on the front panel of the instrument. A scan button is then pressed to start a programmable number of rapid scans of each of the five channels. The electrical signals from the five photodetectors are amplified, converted to digital form and a self contained microprocessor automatically calculates the total column amounts of ozone and water vapor, aerosol optical depth at 1020 nm and the irradiance at each wavelength. This instrument derives total ozone column from measurements of three wavelengths in the UV region. As in the traditional Dobson instrument, the measurement at an additional third wavelength enables a correction for particulate scattering and stray light. Also the ozone calculations based on a single pair of wavelengths show air mass dependence effect. With the use of three wavelengths in Microtops-II air mass dependence correction is effected and measurements are reliable up to an air mass of 3.8. The instrument gives three values of total ozone: $O_3(\text{corr})$, the corrected ozone column in Dobson

Units, ozone column based on the ratio of channels 1 and 2 and that based on the ratio of channels 2 and 3. In this study $O_3(\text{corr})$ value is taken for all further analysis and interpretations.

[7] Observations were made from sunrise to sunset on all days when the sky was clear, at 10 minute interval during the periods following sunrise and preceding sunset and at 30 minute interval throughout the rest of the day. Observations are also made on partly cloudy days mostly in the months of March–May whenever the field of view to the Sun is clear of clouds. An important advantage of Microtops-II when many clouds are present is that an ozone measurement can be made during a few seconds when the Sun is open. Since a Brewer must perform a mechanical scan of wavelengths, it requires more time to conduct an ozone measurement and may not function as well when clouds are frequently blocking the Sun [Morys *et al.*, 2001]. Thus on each clear-sky day about 25 to 30 measurements of total ozone spread over the daytime are made. It was not possible to operate the ozonometer throughout the daytime during the southwest monsoon months of June, July, August, and September due to persistent cloud presence and rain and even if it was operated, it was only for a small duration/part of the day. The average of such data may not be truly representative of the day's average and hence such data has not been considered here. Thus there is a data gap during these 4 months. The observation site, that is the Institute campus, is located almost on the western edge of the urban city of Pune.

[8] Pune city is one of the fastest growing urban centers. It is situated about 100 km inland on the west coast of India, on the lee side of the Western Ghats. To the north east of the main urban area lie the industrial complexes. The current human population of the city would be about 2.8 to 3.0 million and roughly 450 thousand motorized vehicles of all types move on the city roads. The prevailing winds over the location are predominantly Westerly and South-westerly during the summer and southwest monsoon months of March–September and during the winter months (December–February) the Westerlies tend to become weak and may occasionally turn Easterly. Surface meteorological measurements show that daily maximum temperature is highest in the month of April followed closely by May. Higher relative humidity is observed in the monsoon season (June–September) and very dry surface conditions prevail during winter months.

[9] Ozonometer observations of total column ozone collected at this station on 575 mostly clear-sky days during the 5 year period from May 1998 to May 2003 have been used in this study to examine the diurnal, monthly and seasonal variations in total ozone at a tropical urban station. The total ozone measurements obtained from this ground-based instrument have been compared with those simultaneously obtained by Earth Probe-Total Ozone Mapping Spectrometer (TOMS) of NASA over the Pune latitude/longitude. The results are presented and discussed in the following sections.

3. Results and Discussion

[10] Ozone is a fragile atmospheric chemical that is influenced in many ways by its sources, sinks and chemical

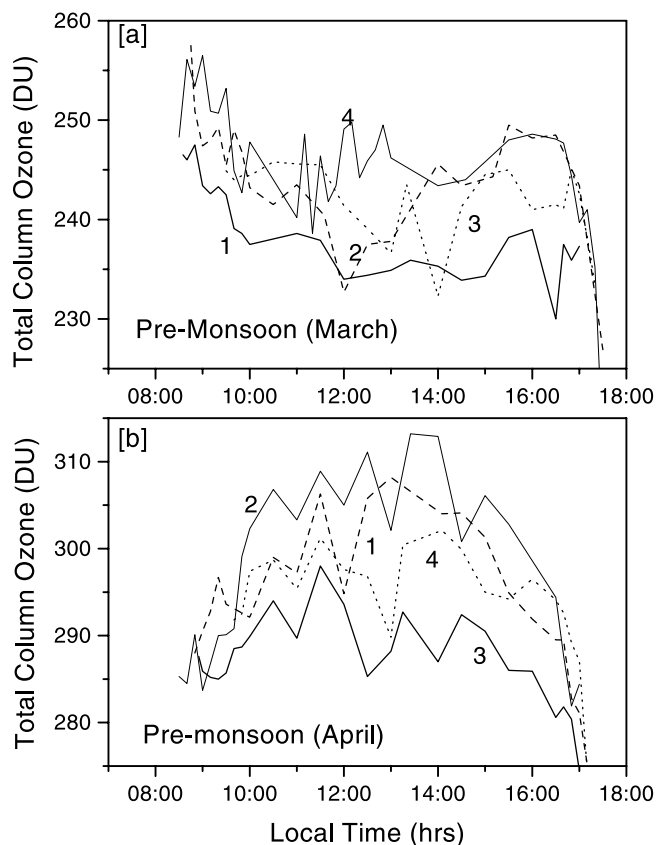


Figure 1. Diurnal (daytime) variation of total column ozone on 4 typical clear-sky days during the premonsoon months of (a) March (1, 17 March 1999; 2, 30 March 1999; 3, 7 March 2000; 4, 6 March 2002) and (b) April (1, 9 April 1999; 2, 13 April 1999; 3, 21 April 1999; 4, 3 April 2000).

reactions. As a result, ozone exhibits significant variation in space and time. Chemical reactions involving ozone formation and removal occur within a timescale of a few hours. An important way of understanding the dynamics of ozone is by examining its diurnal pattern (<http://capita.wustl.edu/NESCAUM/Reports>). As mentioned in the previous section, total column ozone observations have been collected by the authors at this station from around 0730 to 1730 LT. So it is possible to investigate the nature of diurnal/temporal variations in total ozone in the daytime and how it behaves under different meteorological conditions or in different seasons. A significant diurnal oscillation has been reported in the surface level ozone by *Khemani et al.* [1995] and *Shyam Lal et al.* [2000].

[11] A look at the available data suggests that the diurnal pattern is distinctly different in different seasons with reasonably good repeatability in a particular month or season. So a few typical examples of observed diurnal patterns are shown here. Almost all the ozone values correspond to relative air mass less than 3.8 with few exceptions. Figure 1 shows the variation of total ozone during daytime on 4 typical clear-sky days in the premonsoon month of March (Figure 1a) and on 4 clear-sky days during the month of April (Figure 1b). It can be seen that the diurnal patterns are different. During March,

ozone values are already high by the time observations are started in the morning (around 0800 LT) and they begin to decrease slowly till sunset hours. On some days significant short-term fluctuations have been noticed as on 6 March 2002. During the month of April, ozone values are smaller in the morning hours, start increasing and reach a maximum around noontime. They start decreasing after 1500 LT till sunset. It is further noticed that ozone values throughout the daytime are generally higher during the month of April compared to those in the month of March. Also the day-to-day variability in total ozone during the month of April is higher (Coefficient of variation is 6.9%) compared to that in March (C.V. is 5.6%). The expected diurnal variation of ozone has a general pattern. As soon as the Sun rises, ozone too rises, especially at rural sites. Maximum concentration is reached around 1400 LT. Around sunset time ozone concentration drops rapidly as production stops and the produced amount is surface deposited. Thus the temporal variations in Sun photometer-derived total ozone during the month of April seem to follow the expected general pattern. Figures 2a and 2b show the diurnal variation of total ozone at this site during winter (December–February) and during postmonsoon (October–November) seasons respectively. The diurnal

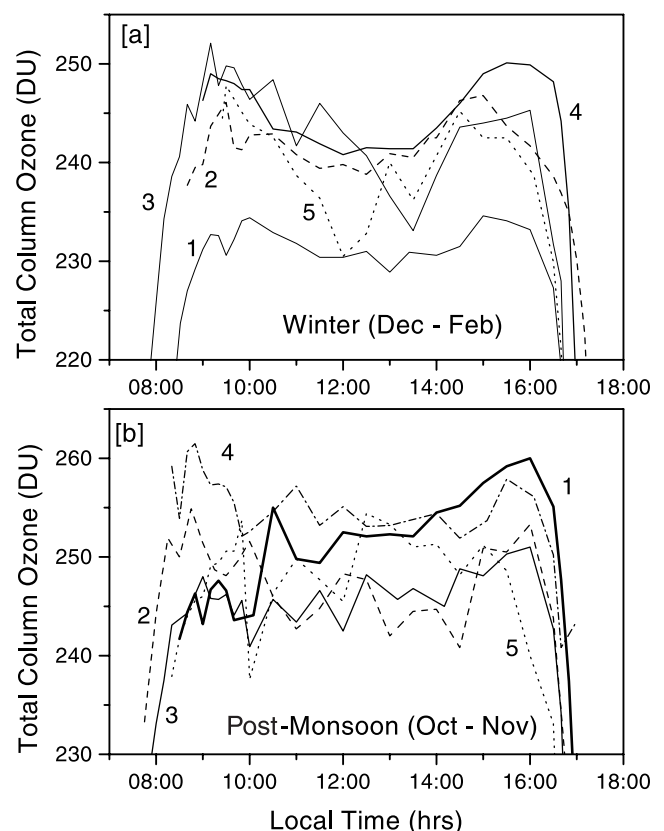


Figure 2. Diurnal (daytime) variation of total column ozone on 5 typical clear-sky days during (a) the winter season (1, 21 December 1998; 2, 27 January 1999; 3, 16 December 1999; 4, 3 January 2000; 5, 6 December 2001) and (b) the postmonsoon season (1, 1 November 1999; 2, 28 October 2000; 3, 13 November 2000; 4, 23 October 2001; 5, 29 November 2001).

variation in winter is very much different from that observed during the premonsoon summer season. Here the total ozone increases rapidly from 0800 LT onward and reaches a peak around 0900 LT. From 1000 LT, ozone values start decreasing slowly till noontime. There is a second peak in the afternoon hours around 1600 LT and after that ozone values fall rapidly. Thus the diurnal pattern during winter shows two maxima, one in the morning period and the other in the evening period, with a distinct broad minimum during noontime hours. Similar feature in diurnal pattern in surface ozone has been reported by Singh *et al.* [1997] for the urban environment of Delhi, India. Owing to strong nocturnal inversion conditions especially in the winter season, at locations close to urban centers, stagnation of surface origin pollutants occurs. The low mixing heights and low ventilation coefficients in winter [Raj and Devara, 1992] can cause increase in ozone concentration levels in the boundary layer during the morning hours. After sunrise, the nocturnal inversion layer lifts and mixing height increases which causes dilution of ozone. This could be one of the reasons for observation of a morning time peak followed by a minimum during noontime in total ozone in winter. Further, the morning time rush hour when vehicular traffic in the urban area increases, may also be another reason for enhancement of ozone concentration in the lower troposphere during the morning hours. The second maximum in the afternoon in the winter season coincides with the evening rush hour of traffic. Here it is assumed that the temporal variations in boundary layer and lower tropospheric ozone content which have surface origin, contribute significantly to those in total column content.

[12] The diurnal variations in total ozone during post-monsoon season (October–November) shown in Figure 2b are nearly same as that observed during winter but the noontime minimum is not prominent. In fact, on some days, ozone is almost constant from morning hours and only shows an increase during the evening hours (around 1600 LT) before decreasing rapidly in the sunset hours.

[13] Thus the distinctly different diurnal patterns observed during different seasons reflect the fact that the ozone and its diurnal cycle are strongly influenced by the meteorological/environmental conditions and human activity. The evolution of the convective boundary layer during daytime and the stable nocturnal boundary layer have a marked impact on the ozone and its precursor gases especially in the lower troposphere which in turn influence the total column content. The different diurnal patterns could be due to a complex combination of various processes.

[14] As mentioned above, there are about 25 to 30 observations of total ozone on each day, spread over the whole day from sunrise to sunset. Daily average total ozone values have been computed for all the available 575 days of observations during the 5 year period from May 1998 to May 2003. The daily mean values of total ozone ranged from 210 Dobson units (DU) to 310 DU during the above period. The large range of ozone values is mainly due to the predominant seasonal variation shown in the later part of the paper. The overall daily mean total ozone for the entire 5 year period is computed to be 254.25 DU with a standard deviation of 18.9 DU. Compared to parameters like aerosol optical depth and precipitable water measured simultaneously with the

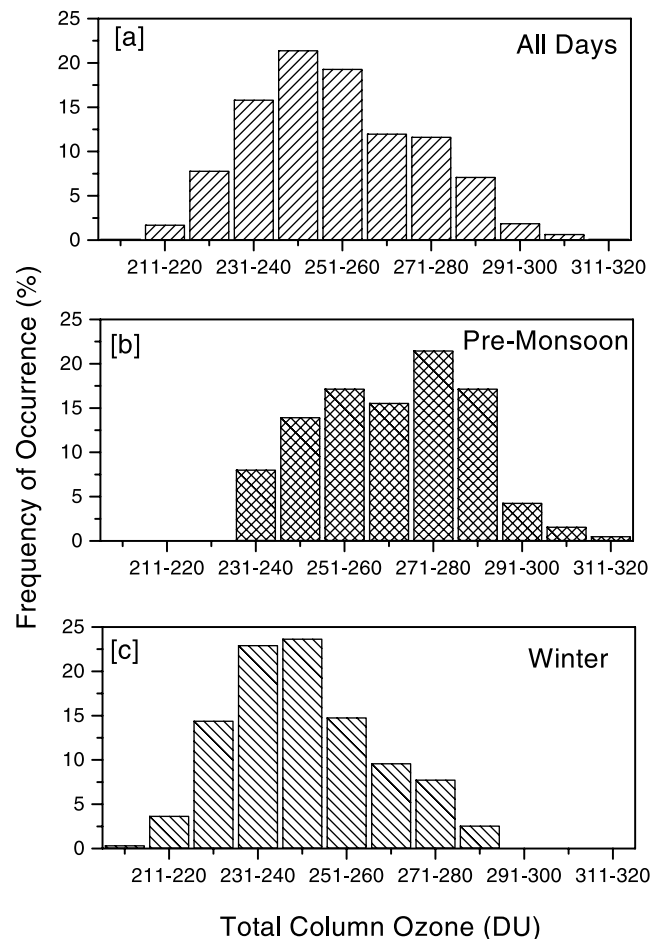


Figure 3. Frequency distribution of daily mean total ozone values for (a) all the days, for (b) the premonsoon season, and for (c) the winter season during the period May 1998–May 2003.

same Sun photometers, total ozone measured at this location showed relatively smaller day-to-day variability of 7.4%. This shows the good overall reproducibility in the total ozone measurements. However, diurnal variability on cloudy sky days is almost 20 DU higher compared to that during clear-sky days. This is confirmed with the increased variability in simultaneously observed aerosol optical depth measurements made during cloudy conditions.

[15] All the available daily average ozone data is taken and grouped into 10 DU wide class intervals between the ranges 201 and 320 DU to compute the occurrence frequencies. Figure 3a shows the percentage frequency distribution of total ozone on all the days. A near normal frequency distribution is obtained with the most probable value of total ozone at this station being in the range 241–250 DU. Similar frequency distributions for the ozone data separately for premonsoon months (March–May) and winter (December–February) is computed and shown plotted in Figures 3b and 3c, respectively. The range of total ozone during premonsoon is between 230 and 320 DU with a most probable value of ozone in the range 271–280 DU. The ozone content range during winter was on the lower side from 201–290 DU with the most frequently occurring value

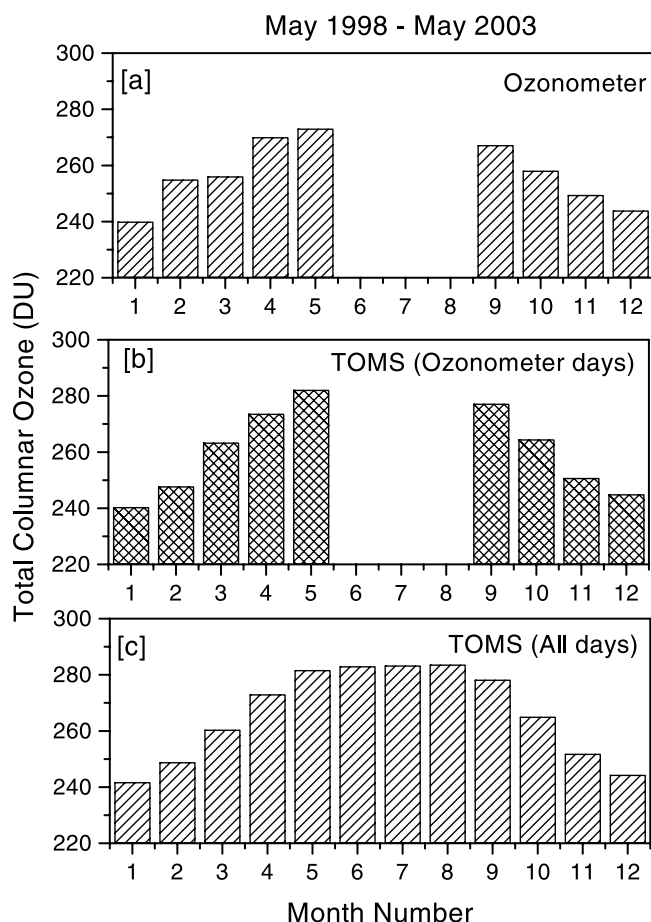


Figure 4. Monthly mean total ozone obtained from (a) the ozonometer and (b) the Total Ozone Mapping Spectrometer (TOMS) on ozonometer observational days and from (c) TOMS on all days.

in the range 241–250 DU. Thus the aforementioned analysis shows that the most probable value of total ozone (241–250 DU) is slightly lesser than the overall mean value (254.25 DU) at this station. Further there is a predominant seasonal variation in the ground-based ozonometer-derived total ozone with a higher range of values during premonsoon summer season compared to those during winter season.

[16] From the daily mean total ozone data, monthly means have been computed for the whole 5 year data to examine the intraseasonal variations and the same is shown plotted as histograms in Figure 4a. One can see the month-to-month change in the total ozone. As mentioned before, very few or no data was available for the southwest monsoon months of June–September, hence the gaps in the plot. It is seen that minimum mean total ozone (240 DU) was recorded during the month of January and it starts building up gradually thereafter. Maximum ozone values (273 DU) over this station are obtained for the month of May. Dobson total ozone data for Pune [Londhe *et al.*, 2003] also shows an identical seasonal variation with maximum in May (282 DU) and minimum during January (245 DU). Their monthly mean data for the period 1981–1998 has been compared with the present Microtops-II

ozone data and it was found that the deviation among the two measurements was within 3%. However, comparison of the data for the same period on daily scale can give more useful information. Daily data of total ozone measured by TOMS from NASA Earth Probe satellite mission for the Pune latitude/longitude for the same 5 year period (May 1998–May 2003) has been picked up from global ozone data sets available on website (<http://toms.gsfc.nasa.gov/eptoms>). From the data on all available days, monthly means of total ozone have been computed and shown plotted in Figure 4c. The month-to-month increase in total ozone from January onward follows closely the observations made with the ground-based ozonometer at Pune. Monsoon months from June to September showed a near constant broad maximum with very little variation. Figure 4b shows the monthly mean ozone from TOMS for only those days on which Pune ozonometer data is available. There is a good correspondence in the month-to-month variation in total ozone from the ground-based measurements and the satellite observations. The coefficient of variation in ozonometer-derived total ozone on monthly scale ranged between 4.6% and 9.2% (corresponding to standard deviation range of 12 to 20 DU), showing again that the variability in ozone is relatively less.

[17] On a seasonal scale, the average total ozone at Pune for the winter season is 246 DU and that for premonsoon/summer months it is 265 DU. Thus the difference of 19 DU between summer and winter seasons is about 8% of the overall mean total ozone over this station. Whereas, on monthly scale, the difference between the minimum and maximum monthly mean ozone is about 33 DU. In the Northern Hemisphere, a high degree of year-to-year variability and largest absolute changes are reported to be observed during the dynamically active winter (December–February) and spring (March–May) periods [WMO, 2003]. Daily averages of total ozone amount at Arosa in Switzerland (<http://iacweb.ethz.ch/en/research/chemie/tpeter/totozon.html>) have shown larger values in spring (March–May), lower values in fall (September–November) and also large day-to-day variability. The systematic seasonal variation observed in the present study agrees with the above results and these may be primarily due to the general circulation features in the stratosphere. Whereas, the day-to-day variability could be related more to the local meteorological and environmental conditions and to the chemical/dynamical processes taking place in the lower atmosphere. As mentioned before, simultaneously observed aerosol optical depth (AOD) values have also shown a very similar seasonal variation with higher values during premonsoon and lower values during winter season. The maximum mixing depth in the daytime derived from Pune radiosonde temperature profile data [Raj and Devara, 1992] showed that the values are around 2 km in the premonsoon months and between 2 and 3 km in the winter months. The height of the boundary layer or the mixing depth can significantly influence the dispersal of pollutants and ozone precursor gases in the lower troposphere. Surface meteorological observations during the 5 year period show that daily maximum temperature is high during the premonsoon months (36.7°C) and small during southwest monsoon (29.2°C) and winter (30.6°C). Relative humidity is

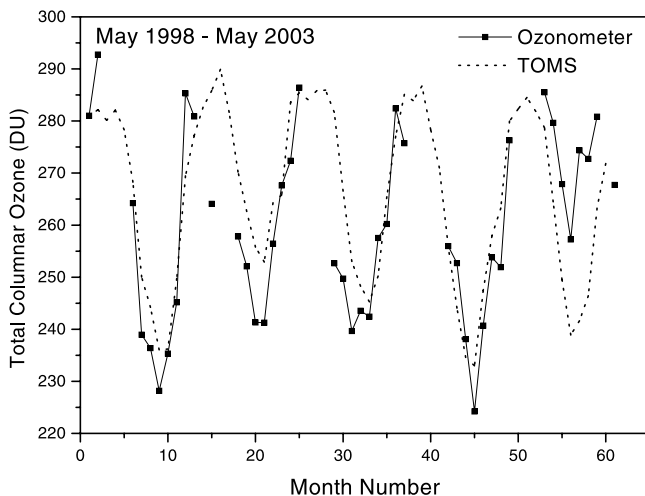


Figure 5. Time series of monthly mean total ozone obtained from the ozonometer and from TOMS (month number 1 corresponds to May 1998).

very low (56%) during premonsoon months and higher during monsoon (83%) and winter months (81%). The large daytime temperatures combined with dry conditions in the premonsoon months may in some way be influencing the lower tropospheric chemistry. Some earlier attempts have been made [Hudson *et al.*, 1995; Hudson and Thompson, 1998; Zienke *et al.*, 1998] to estimate tropospheric content of ozone from TOMS total ozone and to study its seasonal variation. Such studies would provide evidence of relative contribution of tropospheric and stratospheric processes to the diurnal and seasonal variations in total ozone.

[18] To investigate month-to-month variations and long-term changes if any, monthly means of total ozone for individual months derived from both ozonometer and TOMS have been considered. Figure 5 shows these variations from month number 1 (May 1998) to month number 61 (May 2003). The predominant seasonal oscillation in total ozone can be readily seen from the figure with minimum values during winter months of December–January and maximum during summer monsoon months of June–August. It is also noticed that ground-based ozonometer values follow the month-to-month variations in TOMS data very closely. The summer time peak in total ozone does not show any year-to-year change, being nearly constant throughout the 5 year period. However, the winter time minimum shows a significant interannual variation. This could be due to the influence of the quasi-biennial oscillation often reported in total ozone variations. There seems to be no long-term trend in the 5 year data series reported here. However, the data length may be too short to make any conclusive statement regarding the long-term trend.

[19] The correlation in the ozonometer-derived total ozone (present study) and that derived from TOMS observations for the same latitude/longitude location is better depicted in the scatter plot shown in Figure 6a where mean ozone values of all the months in the 5 year period have been taken. The correlation is positive and

the coefficient is about 0.72. The correlation between the two data sets seem to be better (0.80) for the data of premonsoon season (Figure 6b) compared to that of winter months (Figure 6c) when the data sets are separated season wise. However, there seems to be a considerable scatter/bias in the present data which needs to be investigated further. As the ozonometer observations are made from ground-based platform, it is felt that the differences in the above comparisons with the satellite data could be due to the large variability in the surface level ozone concentrations at a location close to urban center. Also the highly variable tropical climate, weather patterns, cloud conditions and local sources may play a role in the observed low correlations or the differences. Such comparisons always help to better intercompare and intercalibrate the ground-based optical remote sensing instruments with other standard instruments.

[20] The seasonal cycle of total ozone is removed by taking the monthly averages for each month of the year for the entire 5 year period (shown in Figure 4a) and subtracting these from the individual monthly means. The departures in mean ozone thus calculated can be expressed as anomalies and are shown plotted for the 61 month period in Figure 7. A strong intraseasonal oscillation can still be seen. In

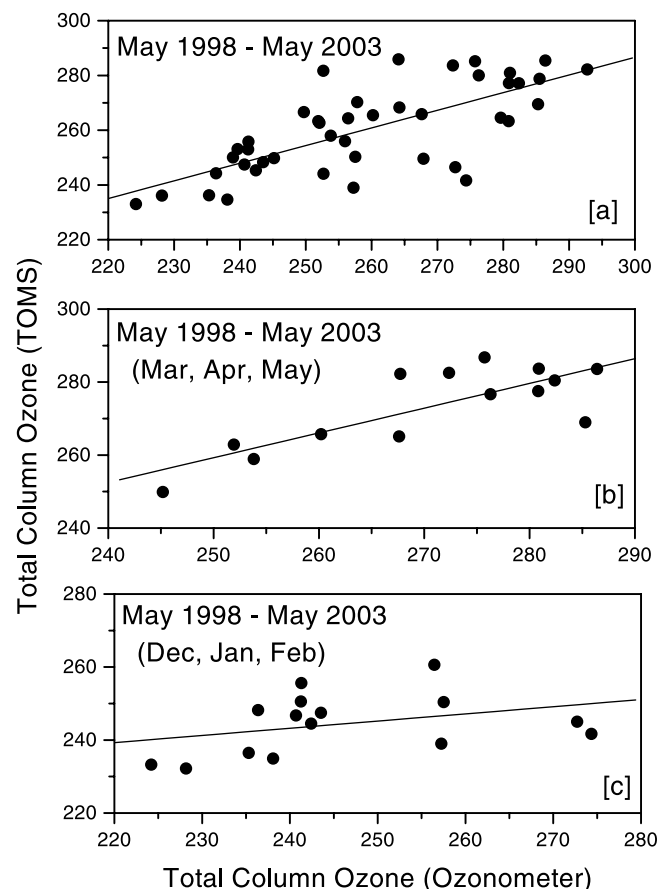


Figure 6. Relation between ozonometer-derived and TOMS-observed total ozone over Pune for (a) all the months, (b) the premonsoon months, and (c) the winter months.

general, winter months show negative departures while the premonsoon months show positive departures. The percentage departures are in the range -7.7 to $+14.3\%$. The departures during the period September 2002–March 2003 are unusually high and positive which needs a closer examination and comparison with other similar data sets for confirmation.

[21] The amount of ultra violet radiation reaching the surface of the Earth is dependant on a variety of atmospheric factors, of which stratospheric ozone is the most important. Enhanced UV levels have been associated with reductions in stratospheric ozone especially over the middle and high-latitude stations. Global ozone measurements from satellite over the period 1979–1993 showed significant UV-B increases at high and middle latitudes of both the hemispheres, but only small changes in the tropics [Madronich et al., 1994]. Such estimates however assume that cloud cover and surface pollution emissions have remained constant over the years. Excessive exposure to UV radiation over periods of time has been linked to increased rates of skin cancer [e.g., De Gruijl, 1999; Mettlin, 2001] (see <http://remus.jpl.nasa.gov/pasadena>), incidence of eye cataracts, to immune system suppression, to reductions in agricultural productivity etc. To examine the relationship between the ozonometer-observed total ozone at Pune and the ground-reaching UV irradiances, daily erythemal dose (kJ/m^2) data has been collected from the web site http://toms.gsfc.nasa.gov/ery_uv/euv.html for the Pune latitude/longitude for all the days in the 5 year period from May 1998 to May 2003. Monthly mean erythemal dose values have been computed. Figure 8 shows the monthly mean ozonometer total ozone and TOMS erythemal dose for the 61 month period. At a first glance both the values appear to vary together but there is a shift of couple of months in their co variation which needs a closer detailed investigation with longer data set.

4. Conclusions

[22] The 5 year Sun photometer data of total ozone at a tropical urban station showed the following.

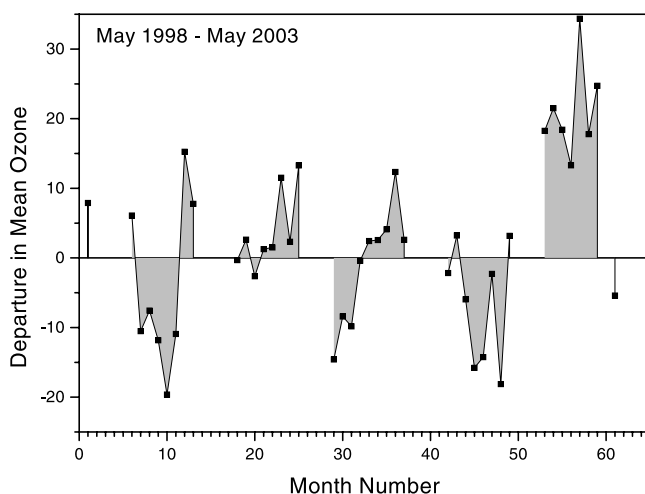


Figure 7. Departures in monthly mean ozonometer-derived total ozone.

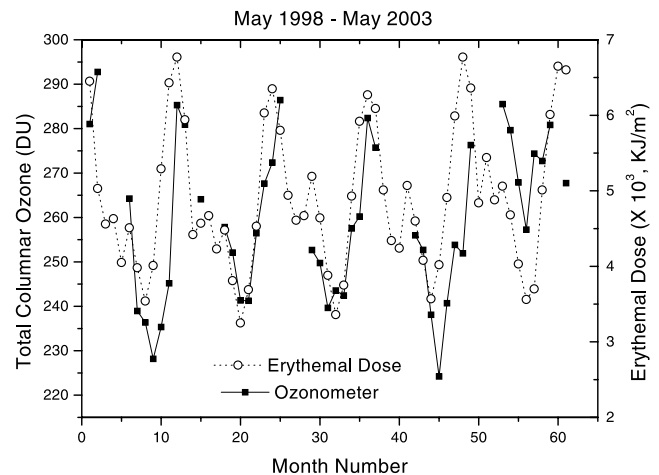


Figure 8. Time series of monthly mean ozonometer-derived total ozone and TOMS-observed erythemal dose for the period May 1998–May 2003.

[23] 1. The diurnal (daytime) variations of total ozone are distinctly different in different seasons and are predominantly due to meteorological/environmental conditions at the surface levels.

[24] 2. The daily mean value of total ozone ranged from 210 to 310 DU, with an overall average of 254 DU. The day-to-day variability is about 7%.

[25] 3. There exists a strong seasonal oscillation in the observed total ozone, with a maximum during premonsoon and a minimum during the winter months, with a difference of about 19 DU.

[26] 4. Ground-based Sun photometer values of ozone compare very well with the simultaneous satellite (TOMS) data.

[27] **Acknowledgments.** Authors are grateful to G. B. Pant, Director, Indian Institute of Tropical Meteorology, Pune, for his encouragement and interest in the present work. The Total Ozone Mapping Spectrometer ozone data of NASA available over their Web site has been used here for comparisons, and the same is gratefully acknowledged.

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