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Temporal and Spatial Patterns of Interannual Variability of Total Column Ozone In Africa from Ground-Based Observations

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Abstract: Total Column Ozone (TCO) measured by the Dobson Spectrometer at Lagos, Nigeria (Lat. 6°27′N, Long 3°24′E) and Cairo, Egypt (30°03′N 31°15′ E) has been analysed in this study. The results give the estimate of the long term annual and seasonal trends from January 1994-December 2004. It also provides statistics of means and variability on temporal and spatial scale. Using linear regression, results show that the TCO increases at an average of 1.34 DU/yr (or 0.50%/yr) in Lagos and 0.37 DU/yr (or 0.13%/yr) in Cairo. On the average an increase of 5.5 and 1.4% in tropospheric total column ozone was recorded in Lagos and Cairo, respectively during the eleven year period. The maximum deviation in daily variability of total column ozone were 38 DU (12.5%) and 91 DU (23.9%) while the minimum deviation were -45DU (-20.1%) and -68.9DU (-31.0%), respectively for both Lagos and Cairo. The range of the seasonal change in Lagos represents 9.2% of the mean TCO value with maximum in August while its 12.1% in Cairo and maximum recorded in March.

Key words: Troposphere, ozone content, photochemical, anthropogenic, biomass burning, trends

INTRODUCTION

Tropospheric ozone is an important atmospheric oxidizing agent and plays a key role in the chemical transformations of many gases, aerosols and hydrometeors (Logan, 1985; Crutzen, 1987). Ozone is produced within the troposphere by the photochemical oxidation of hydrocarbons, methane (CH_4) and carbon monoxide (CO) in the presence of nitrogen oxides ($No_x = NO + NO_2$) (Crutzen, 1988; Marenco *et al*., 1989) and is also transported down from the stratosphere. Ozone photochemical formation occurs in two main ways:

- A rapid formation from reactive hydrocarbons (urban pollution, biomass burning) close to their source, followed by its mixture within the troposphere.
- A slow and delayed formation (2 or 3 weeks) from less reactive precursors, such as CO and CH_4, during or after their redistribution in the troposphere by horizontal and vertical atmospheric motions (Logan *et al*., 1981).

Biomass burning is considered to be an important source of tropospheric ozone in Africa. Combustion products of biomass burning which includes carbon dioxide, carbon monoxide, methane, nonmethane hydrocarbons, nitric oxide, nitrous oxide are chemically active gases that lead to the chemical production of ozone in the lower atmosphere or troposphere. In the last few decades, a global increase in tropospheric ozone concentration was observed mostly in the Northern Hemisphere which has been attributed primarily to the increase in anthropogenic ozone precursors (Volz and Kley, 1988). This has led several scientists and authorities in many regions of the world to perform continuous observations of the variability in O_3 by means of networks able to measure its concentrations. Recent works by Kambezidis (1995), Raga and Le Moyne (1996) have shown that most severe episodes of photochemical smog and in particular those including O_3, are usually found in highly populated and industrialized areas. Many authors have demonstrated that pollution due to photochemical oxidants should be considered on the regional scale and not as a problem limited to purely urban areas (Beck and Grennfel, 1994; Rao *et al*., 1997). There are two main reasons for considering O_3 air pollution at regional level; the first is that the transport of primary pollutants from urban and industrialized zones may lead to alterations in the photochemical activity of other zones at some distance from the source of emission (Meagher *et al*., 1998). The second is that meteorological and synoptic conditions, which affect photochemical processes and pollutant transport, tend to affect quite large areas, meaning that the variability of O_3 follows homogeneous patterns in such areas. Cox and Clark (1981) consequently reported the need to take into account such regional patterns when attempting to assess the progress achieved in air quality control strategies instead of analyzing the measurements from individual stations. Examples of studies that have focused on constructing ozone climatology for different regions include (Kirchhoff *et al*., 1991; Diab *et al*., 1996; Thompson *et al*., 2003).

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Satellite based instruments provide the spatial coverage of earth compared with ground based instruments for studying the global distribution, source and sinks, transport, seasonal behaviour and trend of tropospheric ozone. Analysis of tropospheric O_3 trends, especially O_3 near the surface, has suffered from scarcity of long-term measurement data especially in Africa. The most extensive measurements are carried out in Europe and North America, both at the surface and with ozonesondes (Logan, 1994). In this study we report the results of a long-term study of ozone over a period of eleven years performed in Lagos and Cairo. The magnitudes of maxima and minima, mean profiles averaged by season or year, together with some measure of variability about these means, will be analyzed.

MATERIALS AND METHODS

For this research, Dobson network data for the period January 1994 to December 2004 were retrieved for Lagos and Cairo. The stations (WMO station numbers 317 for Lagos and 152 for Cairo) are operated by the Nigeria Meteorological Agency and the Egyptian Meteorological Agency, respectively. The World Ozone and Ultraviolet Radiation Data Centre (WOUDC) is one of six recognised World Data Centres which is part of the Global Atmosphere Watch (GAW) program which in turn is part of the World Meteorological Organization (WMO). Measurements of ozone are collected since 1960 and available through the WOUDC website (http://www.tor.ec.gc.ca/woudc/woudc.html). The instruments are described in detail by Bramstedt *et al.* (2003) which is herein describe briefly. The Dobson spectrophotometer is a quartz double monochromator which measures the relative intensities of pair wavelengths in the Huggins ozone absorption band (300-350 nm) from which total ozone in the atmosphere can be deduced. The first Dobson spectrophotometer was developed in 1927 by G. M.B. Dobson (Dobson, 1931). Since then Dobson spectrometer plays an important role in routine measurements of total ozone. Dobson instruments need an accurate calibration, limiting historical records to an accuracy of 5-10% (Grant, 1989). Since the mid-1970s, virtually all instruments in the Dobson network including that of Lagos and Cairo are regularly calibrated with the reference standard Dobson spectrophotometer M83 located in Mauna Loa, Hawaii (Bramstedt *et al.*, 2003). The relative uncertainty is estimated to be 2% (Basher, 1985).

Monthly and yearly means are estimated based on the daily measured values, assuming that they are representative of daily means. To examine the trends for each station, linear regression analysis from time series line plots of the daily, monthly and yearly TCO were performed. The TCO variability in the stations was investigated by using the Standard Deviation (SD), Standard Error (SE) and the Coefficient of Relative Variations (CRV). The CRV is simply estimated as CRV = 100*SD/mean which is useful for comparing relative variability of two or more variables whose means are dissimilar (Chen and Nunez, 1998).

RESULTS AND DISCUSSION

Figure 1 a and b shows the daily TCO at Lagos and Cairo from January 1994 to December 2004. Also shown in Fig. 1 is the 12-point binomial smoothing to successive daily values so as to reduce the complexity of the variations. The figure indicates that despite considerable short term variations, there exits a seasonal change for the period of analysis at both stations. The estimated mean TCO were 268±14 and 291±21 DU for Lagos and Cairo, respectively. This gives a Coefficient of Relative Variations (CRV) of 5.22 and 7.22%, respectively. The maximum and minimum TCO in Lagos during the period were 306 and 223 DU, respectively, giving a range of 83 DU which represent 30.98% of the mean value. However in Cairo, the maximum and minimum during the period 1994 -2004 were 382 and 222 DU, respectively, with a range of 160DU also representing 55% of the mean value. To establish a long term trend based on the daily observations, linear regression is performed on the data, resulting in a trend of 0.00295DU/day or 1.08DU/yr (0.41%/yr) in Lagos and compared to 0.00037DU/day or 0.14DU/yr (0.05%/yr) in Cairo. This is in agreement with Chandra and Stolarski (1991) that reported similar positive correlation at the equator. The result can further be compared with an average increasing trend of 0.14±0.04 DU/yr (or 0.9±0.3%/yr) over the eastern Pacific Ocean (Kim and Newchurch, 1996).

Figure 2 presents the variation in daily average total ozone expressed as percentage deviation from the mean, where the mean is based on data for the 11-year interval. The maximum increase and decrease in the daily variation in Lagos were 38 DU (12.5%) and -45DU (-20.1%), respectively whereas the values were 91DU (23.9%) and -68.9DU (-31.0%) in Cairo. The standard deviation of the variation which gives the measure of the mean change from day to day is 9.5 DU (3.56%) for Lagos and 12.5 DU (4.29%) in Cairo.

The monthly averages of the TCO from January 1994 -December 2004 is shown in Fig. 3a with a clear seasonal change. This is a consequence of varying influences which include large scale dynamics, vertical mixing and redistribution through convection, advection of pollution

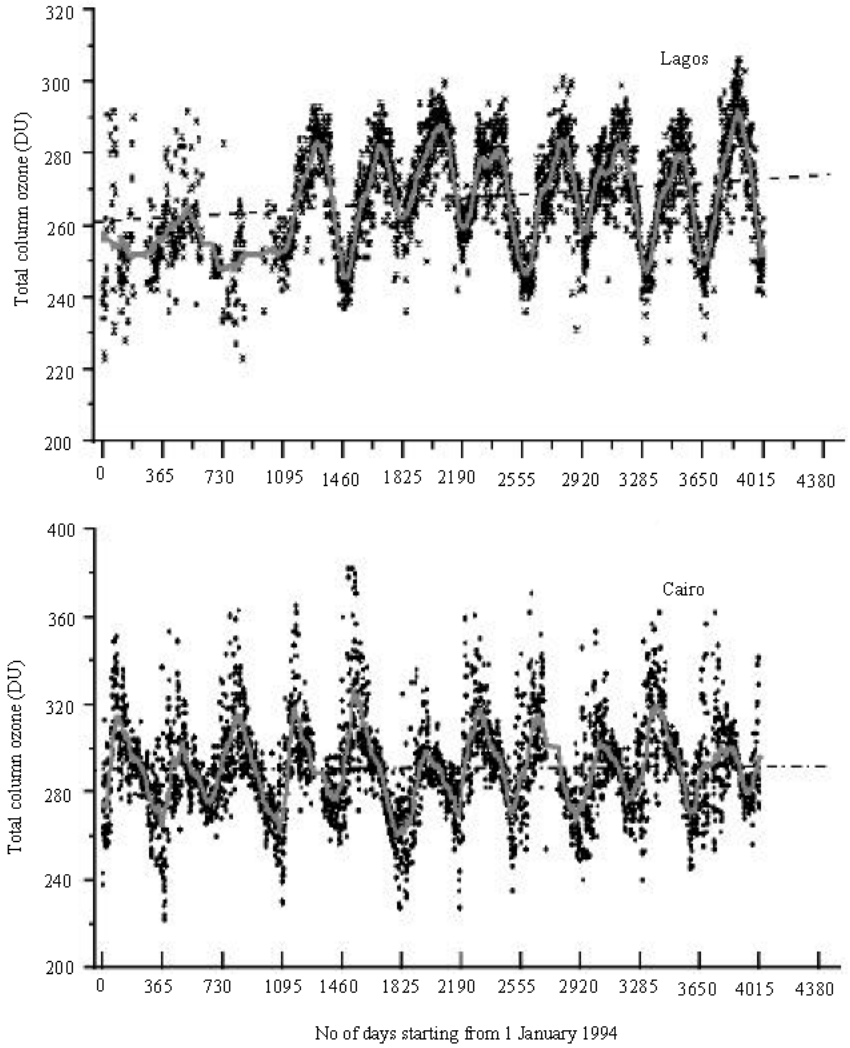

Fig. 1: Daily tropospheric Total Column Ozone (TCO) content in Dobson unit recorded at Lagos and Cairo from January 1994-December 2004. A 12-point smoothing shown in grey is applied to the daily average values

with ozone precursors, biomass burning and biogenic sources (Thompson *et al*., 2003). Linear regression of the monthly observations in Lagos yields a trend of 0.10DU/month (1.20DU/yr). The maximum and minimum monthly total column ozone values during the 11 years period at Lagos were 244 and 278DU recorded in April 1996 and October 2004, respectively. The variations of monthly deviation from average expressed in DU are illustrated in Fig. 3b which shows a short term disturbance with clear seasonal variability. The largest and smallest monthly deviations recorded in Lagos were 28DU (11%) and -35DU (-13%), respectively. In order to identify various seasonal changes of the day-to-day variability of the TCO normalised monthly standard deviations by their means based on the daily value for all the months were estimated and shown in Fig. 3c. The largest seasonal variability is obvious in December-February and smallest in the summer months of June-July. In a similar manner Fig. 4a illustrates the monthly averages of the TCO at Cairo. Figure 4 yields a slight positive trend of only 0.012DU/month (0.14DU/yr). The maximum increase and decrease in the monthly deviation variation shown in Fig. 4b were 43DU (15%) and -31DU (-11%), respectively in Cairo. Figure 4c shows that on the average, CRV from the mean of the monthly mean TCO are largest in May and minimum in September. The mean monthly values of TCO in Lagos and Cairo were 265.3±13.4 and 291.1±15.3 DU, respectively.

To further quantify the seasonal variation, monthly means of the TCO from 1994-2004 were estimated for

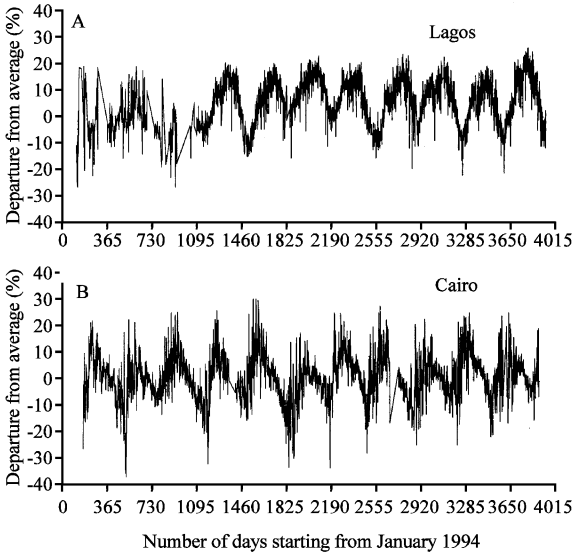

Fig. 2: Variation of daily average total ozone expressed as percentage deviation from the mean at Lagos (a) and Cairo (b) from January 1994-December 2004

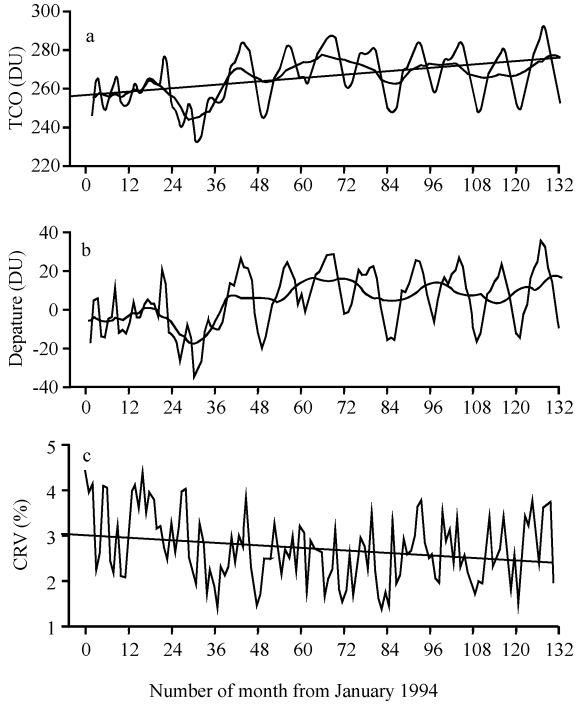

Fig. 3: Statistics of monthly total column ozone at Lagos (a) monthly average TCO estimated from daily values (b) monthly departure from average (c) the coefficient of relative variation. Also shown in grey lines are the 12-point smoothing applied to the monthly average values

Lagos and Cairo as shown in Fig. 5a. The maximum and minimum monthly mean of TCO were 276 DU in August and 252 DU in January. The range of the seasonal change amount to 24 DU which represents 9.2% of the mean value measured in Lagos. The ozone amount being higher in summer than in winter is assumed to be due to photochemical ozone production from increasing anthropogenic emissions of trace gases. In Cairo the maximum and minimum mean monthly TCO recorded were 308 DU in March and 273DU in November, respectively. This is in agreement with an ozone enhancement in equatorial North Africa observed in March which is the biomass burning season as reported by Kim *et al*. (2001). The difference between the maximum and minimum represents 12.1% of the mean value. Figure 5(b-c) represents the monthly means SD and the related CRV recorded in both stations.

The annual means during the period 1994-2004 is computed for both Lagos and Cairo as shown in Fig. 6a. The annual average values during the 11year observation were 265.0±6.8 DU and 291.4±3.4 at Lagos and Cairo, respectively. Figure 6 shows that the annual mean were lower during the periods 1994-1996 and gradually increases after 1996 in both stations. This could have

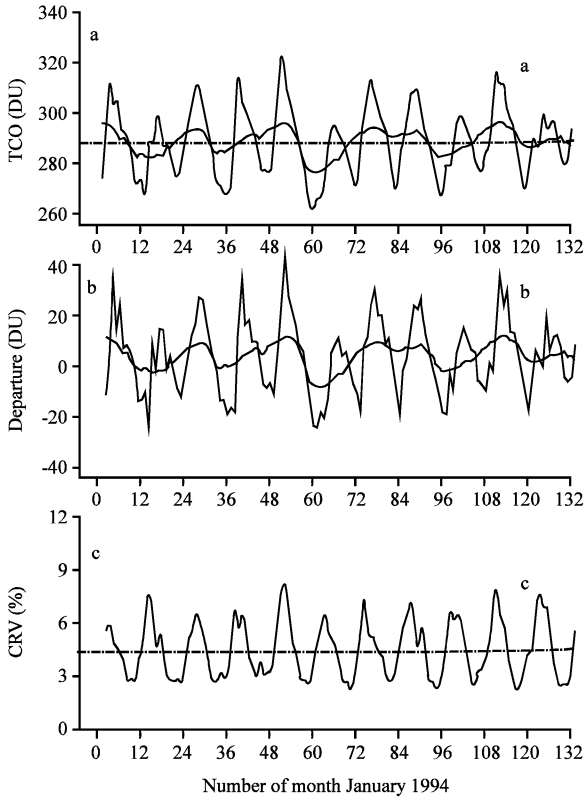

Fig. 4: Same as in Fig. 3 above but data points are for Cairo

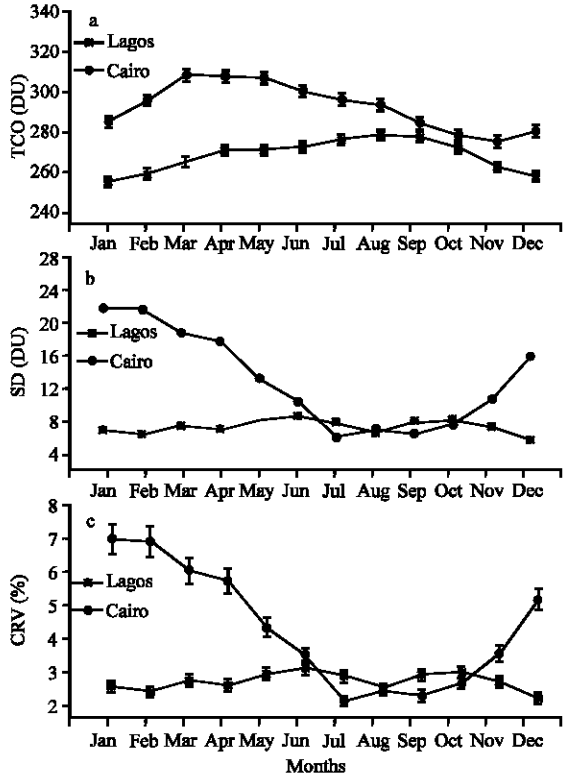

Fig. 5: Monthly variation of tropospheric total column ozone in Lagos and Cairo. Also indicated is the error bar that represents±2% standard deviation. (a) seasonal mean TCO (b) seasonal standard deviation (c) seasonal coefficient or relative variation

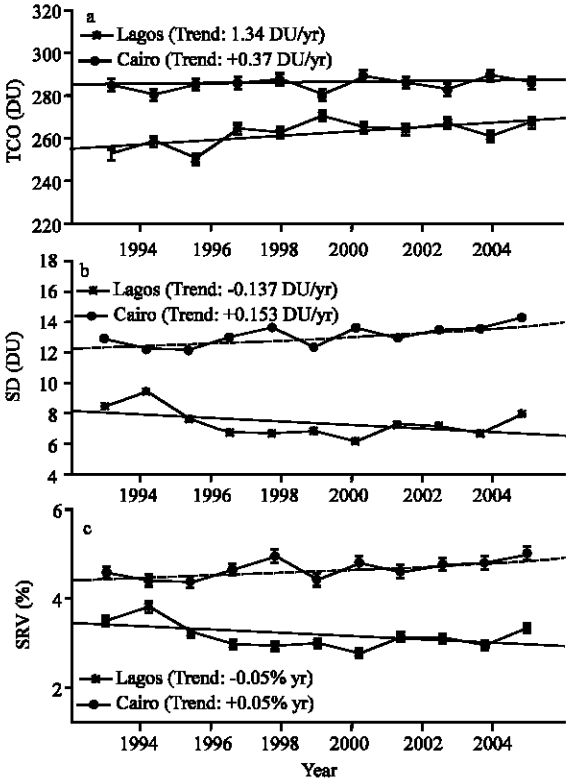

Fig. 6: Temporal variation of tropospheric total column ozone in Lagos and Cairo. Indicated is the error bar that represents±2% standard deviation and the fitted linear trend lines. (a) annual mean TCO (b) annual standard deviation (c) annual coefficient or relative variation

been enforced by the 2-4% lower ozone values during 1991-1993, part of which might be related to the effects of the Mt. Pinatubo eruption and might also be due to the strong Quasi-Biennial Oscillation (Bojkov and Fioletov, 1996). Figure 6a gives an estimated trend of 1.34 DU/yr or 0.50%/yr in Lagos and 0.37 DU/yr or 0.13%/yr in Cairo. The trend implies that on the average the lower troposphere gained 5.5 and 1.4% of the total ozone during the eleven year period at Lagos and Cairo, respectively. Similar increases in surface ozone measurements, at some stations in Germany, show a long-term linear increase of 1-3% /yr (Feister and Warmbt, 1985). The trends for Lagos and Cairo can further be compared with the global decreasing trend of 5 and 6.5% on average for middle and high latitudes during the last 15 years (Bojkov, 1995). The long-term ozone increase cannot be fully explained by a change in the ozone transport from the stratosphere, but should rather be due to photochemical ozone production in the troposphere (Logan *et al*., 1981). The photochemical ozone production by oxidation of CO, CH_4 and Nonmethane Hydrocarbon (NMHC) in the presence of NOx and solar UV radiation (2, -< 400 nm) occurs under the condition of extreme pollution within the cites. This may be attributed to the rapid industrial development, urban-industrial emissions growing energy consumption, increasing traffic density and altered technical practices (e.g., higher combustion temperatures with fossil fuel burning) (Diab *et al*., 2004). The anthropogenic emissions of CO, NO_x and NMHC have increased in highly developing cites like Lagos and Cairo within the past decades. If the surface ozone trends continue, the ozone concentration as well as its seasonal amplitude will increase significantly unless preventive measures is implemented as to reduce or to keep at a fixed level the anthropogenic emissions of NOx, CO and hydrocarbons. The increasing ozone amounts can have a number of unfavourable effects including enhanced losses in agriculture and forestry, adverse effects on human health and climatic effects in this region. Thus it seems appropriate to tackle the following tasks in atmospheric

science research in Africa: To start or continue regular observations of the ozone concentration near the surface, in the boundary layer and in the free troposphere in many parts of Africa with the required frequency so as to provide evidence for the real spatial extent of the ozone increase;

- To start or continue regular measurements of trace gases which affect the ozone distribution in the region.
- To improve the instruments' reliability and accuracy and calibration so as to obtain consistent and reliable data.
- To set up emission inventories of trace gases. To develop, apply and validate models which simulate the effect of anthropogenic emissions on the ozone distribution in the boundary layer.

The validated model results can then be used as scientific background for necessary emission reductions of specific trace gases and for the assessment of trans-boundary transport. Annual mean standard deviations and CRV based on the monthly data are estimated and plotted in Fig. 6 b and c. It is interesting to note that as the variations reflects the month-month variation; The two parameters show a decreasing trend for Lagos while an increasing trend is observed in Cairo. A linear fit for SD and CRV in Lagos gives an estimated trend of -1.37DU/yr and -0.0519%/yr and 0.1528 DU/yr and 0.051%/yr for Cairo, respectively.

CONCLUSION

This study has analysed surface ozone measurements from 1994 to 2004 which have been set up in Lagos (Nigeria) and Cairo (Egypt). The estimated daily mean TCO were 268±14 and 291±21 DU and coefficient of relative variations of 5.34 and 7.31% for Lagos and Cairo, respectively. The largest and smallest monthly deviations of 28DU (11%) and -35DU (-13%) were recorded in Lagos and 43DU (15%) and -31DU (-11%), respectively in Cairo. The study further shows a long-term significant linear increase of 0.50% yr^{-1} in Lagos and slight increase of 0.13% yr^{-1} in Cairo. The trends are assumed to be due to photochemical ozone production from anthropogenic trace gases and biomass burning in North and sub Saharan Africa. Since the photochemical ozone production arises from the sources of CO, CH_4 and NMHC, it is suggested that the tropospheric ozone concentration will continue to increase, unless the anthropogenic emissions of ozone precursors are kept at fixed levels or are even reduced.

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