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ANALYSIS OF THE TEMPORAL EVOLUTION OF TOTAL COLUMN NITROGEN DIOXIDE AND OZONE OVER NAIROBI, KENYA USING DAILY OMI MEASUREMENTS

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Abstract
Concurrent measurement and analysis of Nitrogen dioxide (NO_2) and Ozone (O_3) are essential for improved understanding of ozone distribution. This study sought to analyse the temporal evolution of total column NO_2 and O_3 over Nairobi using satellite-derived daily data between 2009 and 2013. Seasonality is observed in O_3 distribution with minimum and maximum occurring during the dry and wet seasons, respectively. Additionally, a lag of about a month or two occurs between the onset of a season and corresponding minimum or maximum NO_2 and O_3 concentration. The established association between monthly NO_2 and O_3 is such that, above average concentration of NO_2 is likely to lead to above average levels of O_3 during the same month (r=0.79) and below average levels about 5 months later (r=0.39). The Quasi Biennial Oscillation (QBO) is the main phenomenon behind the oscillating biennial feature exhibited by NO_2 and O_3 interannual trend. The study shows that NO_2 and O_3 are increasing at annual average rates of about 0.27% and 0.46% per year compared to mean values, respectively. Daily variation of both NO_2 and O_3 depicts stagnating trends over the entire period of study. This difference is attributed to the fact that, whereas daily NO_2 and O_3 are influenced by mechanisms that control the slow shift between the dry and wet periods within the course of a year, interannual variability is driven by the differences in each year's general weather conditions.

Key Words: *Evolution, Nitrogen dioxide, Ozone, Total Column, Quasi Biennial Oscillation*

Introduction

Ozone (O_3) and Nitrogen dioxide (NO_2) are very significant species in the Earth's atmospheric chemistry and have an influence on radiative forcing of the climate system (e.g., Herman, 2010). The atmospheric formation, dispersion, destruction, and transport of O_3 are influenced by meteorology and photochemical mechanisms involving various species such as oxides of nitrogen (David and Nair, 2011). For this reason, simultaneous measurements and analysis of O_3 and NO_2 are, therefore, essential for improved understanding of ozone photochemistry and variation.

NO_2 is produced primarily by combustion processes resulting from transportation, industry, and power plants (Seinfeld and Pandis, 2012). Over South Africa for example, Edward *et al*. (2003) were able to identify biomass burning and lightning as the two principal tropical sources of this important O_3 precursor.

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Reactive nitrogen compounds play a critical role in atmospheric chemistry, directly and indirectly controlling ozone production (Finlayson-Pitts and Pitts, 2000). Moreover, NO_2 may also play a significant role in radiative forcing of the atmosphere (Solomon *et al*., 2007). Stratospheric NO_2 shows a diurnal cycle with maximum concentrations around sunset and a seasonal cycle with maxima in summer and larger abundance at midlatitudes and high latitudes than in the tropics (Liley *et al*., 2000).

In the troposphere, nitrogen oxides significantly contribute to poor air quality (Celarier *et al*., 2008). At short-term concentrations exceeding 200μg/m^3, NO_2 is a toxic gas which causes significant inflammation of the airways (WHO, 2006). Tropospheric NO_2 depicts seasonal patterns showing winter maximum (Cede *et al*., 2006). Globally the distributions of tropospheric NO_2 columns are lowest during summer, due to rapid loss by reaction with OH (Martin *et al*., 2003). At any given location, the diurnal variation is the result of a complex interaction between the emission source field, photochemical lifetimes, advection, and the levels of chemical sinks for NOx species (Celarier *et al*., 2008).

The temporal changes in O_3 concentrations, driven partly by changing seasons, have implications on the climate, vegetation and lives on earth. Photochemical reaction of pollutants such as NO_x results in ground level O_3. At levels exceeding 100μg/m^3, O_3 in the air can cause among other human health effects, breathing problems (WHO, 2006). It is an important gas because of its absorption of the biologically harmful ultraviolet radiation, thereby protecting live on the earth's surface.

Both stratospheric and tropospheric O_3 is largely produced naturally through photochemical and chemical reactions (Ogunjobi *et al*., 2007). However, the human-induced depletion of O_3 by Chlorofluorocarbons (CFCs) is more of a concern because the process is rapid. The distribution of O_3 is influenced by seasonal transport from one location to the other as a result of seasonal circulations in the atmosphere. It implies, therefore that O_3 concentration will vary depending on the season of the year. For example, seasonal O_3 variation showing springtime maximum over South Africa have been revealed using ground-based observations (Diab *et al*., 2004) emphasizing the influence of synoptic weather systems. Over equatorial Africa, the lower troposphere exhibits layers of enhanced O_3 during the biomass burning season in each hemisphere (Sauvage *et al*., 2004). The exchange of O_3 between the stratosphere and troposphere also take place on a seasonal scale (Diab *et al*., 2004).

Whereas the station observations of air pollutants concentrations in the past have often been limited in spatial coverage, the more recently available satellite measurements now provide a means to link local production of pollution with regional and global scale transport (Edwards *et al*., 2003). In Africa as a whole, only few studies have illustrated the variability of criteria pollutants due to inadequate ground–based measurements. Moreover, such studies are centered on and around South Africa. Thus, there is sparse documentation on this important research area. Since the South and East Africa where Nairobi is situated have climatic disparity, a distinct difference in their selected air pollutant climatology is expected. This study, using satellite data aims at bridging this important gap.

The aim of this study to analyse in detail, the temporal variation of total column NO_2 and O_3 on daily, monthly and

interannual annual basis over the Nairobi County for the time period of 2009-2013. It relates the temporal evolution of total column NO_2 and O_3 to the climatology of the study area with an intention of establishing the role of the area's varying seasons on their concentrations.

Study Area

Nairobi is situated at 1.17ºS, 36.49ºE in the highlands of the southern part of the country. It is Kenyan's capital city and the country's major economic and administrative centre and is one of the largest and fastest growing cities in Africa. The city has the highest urban population in East Africa, estimated at 3.14 million (KNBS, 2009) living within 684 km^2. At roughly 1660 m above sea level, Nairobi has a fairly moderate climate. The two main seasons are; the long wet season between March and May and the dry season between December and February of the following year. Figure 1 is a map of the study area; showing Nairobi County.

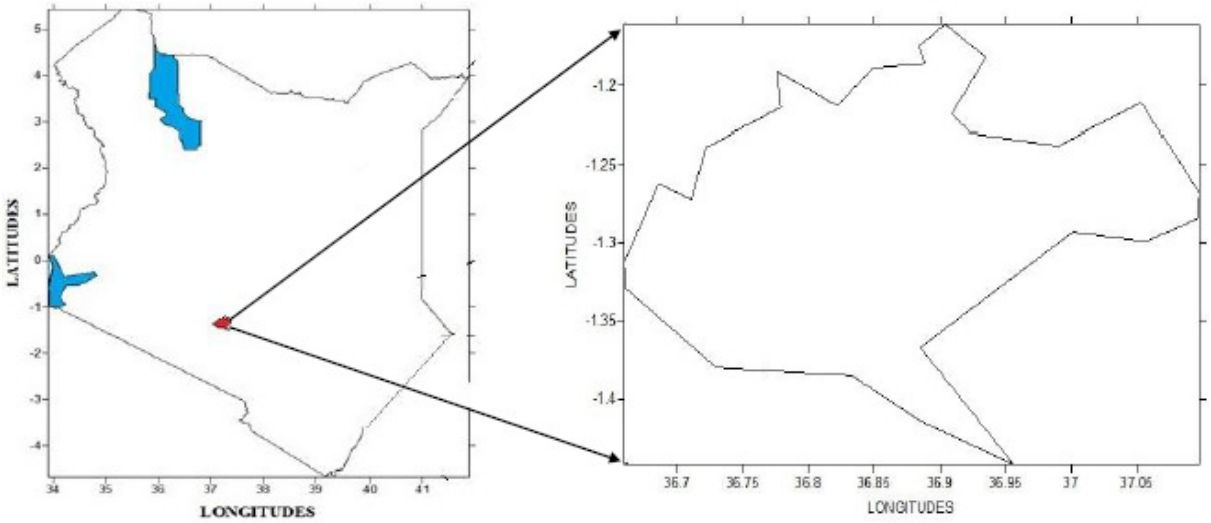

Figure 2: Map of Kenya (right) showing the location of the study area; Nairobi County (left)

Methodology

Data Collection

The data used in this study are daily total column amount nitrogen dioxide (OMNO2e version 3) and ozone (OMTO3e version 3) based on TOMS algorithm from 1 January 2009 to 31 December 2013 over Nairobi. These were obtained using Giovanni interface for visualization and analysis of the Earth Observing System (EOS) Aura Ozone Monitoring Instrument (OMI) version 3 daily level 3 global 0.25° gridded data. OMI is a spaceborne spectroradiometer that flies onboard the National Aeronautics and Space Administration's (NASA) EOS Aura satellite launched in July 2004. OMI employs the concept of measuring the complete spectrum in the ultraviolet/visible/near-infrared wavelength range with a high spectral resolution. Even though TOMS has the advantage that it has a fairly small ground-pixel size (50 km by 50 km) in combination with a daily global coverage, OMI combines the advantage of Global Ozone Monitoring Experiment (GOME) and Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) with the advantage of TOMS, measuring the complete spectrum in the ultraviolet/visible wavelength range with a very high spatial resolution (13 km by 24 km) and daily global coverage. This is possible by using a two-dimensional (2-D) charge-coupled

device (GCD) array detector (Celarier *et al*., 2008).

The OMI's dataset has been extensively validated, and the errors are reasonably well understood (Celarier et al., 2008). OMI total column NO_2 appears to have a negative bias with respect to measurements from ground-based and DS-DOAS (Celarier *et al*., 2008), GOME (Martin *et al*., 2002) and SCIAMACHY (Schneider and van der, 2012). Even with the significant apparent bias, a good correlation is reported, generally >0.6 (Celarier *et al*., 2008). Again, OMI's O_3 data have been found to compare reasonably well with ground-based measurements and other satellite platforms; a globally averaged agreement of better than 1% for OMI-TOMS data and better than 2% for OMI-DOAS data with the ground-based observations.

The data resolution is 0.25° latitude by 0.25° longitude. The readings were extracted from single cell within which the surface station lies with the aid of a simple R-NETCDF program. The data are measured in Dobson Units (DU).

Results and Discussion

Daily Variation

The cyclic nature of both daily NO_2 and O_3 concentration depicts a seasonal phase, exhibiting a wavelength of about 420 Julian days (starting from 1st January, 2009). This implies that shorter than one cycle is complete in a year as shown in Figure 2. The cycles start with a minimum during the period of dry season (December-January), and gradually assume a maximum during the period of wet season (March-May) and cold season (June-August). Minimum and maximum NO_2 and O_3 concentrations over Nairobi are found to occur principally in January and September, respectively. However in Nairobi the period of warm and dry season occurs in December and extends to February of the following year, period of cold and dry occur in June-August while periods of wet and cold seasons occur in March-May (long rains) and October-December (short rains), respectively. This implies a one-month lag between the onset of a season and minimum or maximum NO_2 and O_3 concentration, correspondingly. For the total number of years considered, NO_2 maximum and minimum concentration over Nairobi was 6.9 DU and 1.8 DU, respectively producing an average range of 4.8 DU, which is about 145.45% of the mean value (3.3 DU). On the other hand O_3 maximum and minimum level was 289 DU and 227 DU, respectively producing an average range of 62 DU, only about 24.16% of the mean value (256±6.9). Comparable results (254.35±8.25 DU) have been reported by Ogunjobi *et al*. (2007). It was noted that at no day during the entire study period over Nairobi was ozone concentration below 220 DU minimum mark otherwise known as ozone hole, recorded. Odhiambo et al. (2010) have reported that even though O_3 and NO_2 are within the WHO guidelines over Nairobi, higher levels of NO_2 are detected during the morning and evening traffic peak hours. This clearly shows that motor vehicles are the most probable source of these gaseous pollutants within the city centre.

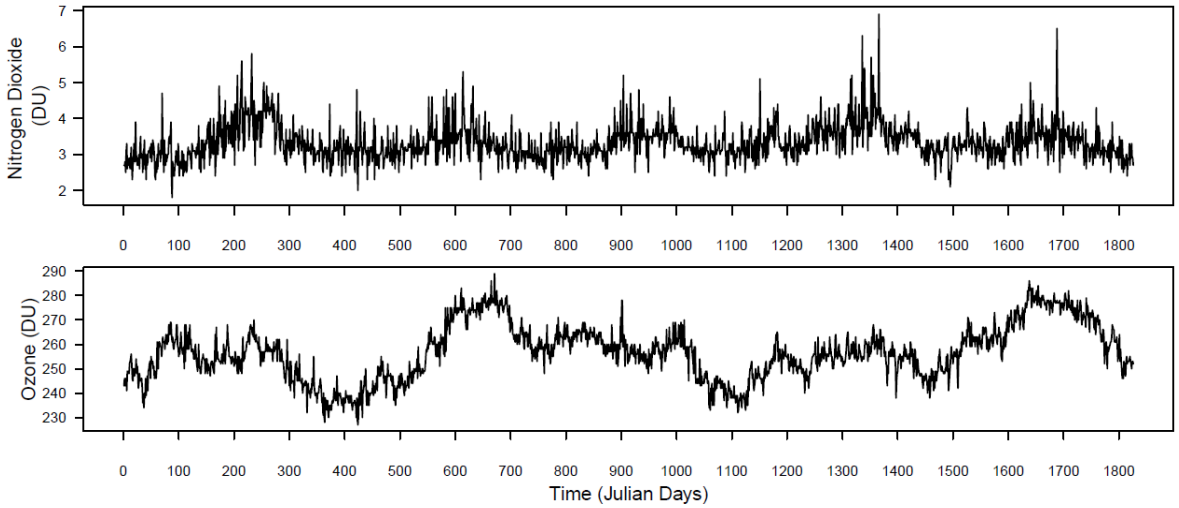

Figure 3: Cyclic nature of both daily total column NO_2 and O_3 concentration over Nairobi

Monthly Variation

The shapes of mean monthly evolution of total column NO_2 and O_3 for the five-year averaged data are as shown in figure 3. Both NO_2 and O_3 concentrations generally grow from January and reach peaks in September which then diminish thereafter to December. Precisely, Ozone depicts a slight peak during the long rains in March-May and a major peak in August-October just before the commencement of the short rains. Bundi (2004) also showed that tropospheric O_3 depicts a maximum during this period over Nairobi. Minimum values are realized in December-February. Similar seasonal patterns have been observed by Ayoma *et al*. (2002) with total O_3 measurements by Dobson instrument. Thomas *et al*. (2003), attributes enhanced total column O_3 and unusually high levels of NO_2 during such periods to severe thunderstorms and extreme rainfall and intrusion of stratospheric air. According to Thomas *et al*. (2003) much of the total atmospheric content of NO_2 is found in the troposphere, due to lightning activity, advection and vertical transport in the thunderstorms from the planetary boundary layer (PBL) to atmospheric levels above clouds. Diab *et al*. (2004) have suggested that similar seasonal springtime tropospheric ozone maximum over South Africa is strongly influenced by non-seasonally fluctuating sources such as urban-industrial emissions because of its proximity to major metropolitan and industrial areas. Seasonal enhancements from biomass burning occur over northern Africa during December-February and over central Africa during June-October (Martin *et al*., 2003).

Despite the similarity in the shape depicted by monthly evolution of nitrogen dioxide and ozone over Nairobi, the disparity in (spike) peakedness of the evolution, particularly the dip is however noted.

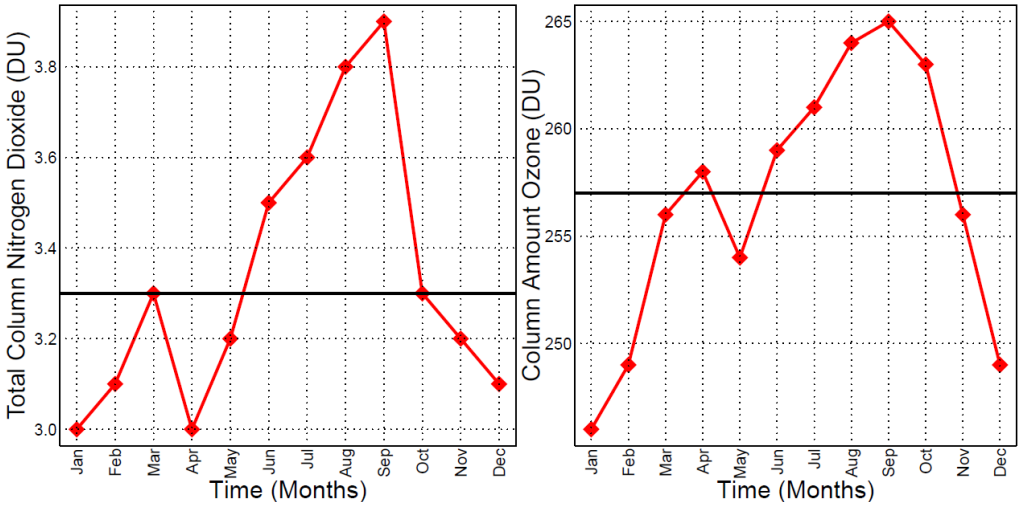

Figure 4: Mean monthly evolution of total column NO_2 and O_3 for the five-year averaged data

The persistence in both the monthly NO_2 and O_3 trends was examined by calculating their autocorrelation functions (ACFs). Sample autocorrelation coefficients measure the correlation between observations at different times (Box and Jenkins, 1976). The set of such autocorrelation coefficients arranged as a function of separation in time is the ACF. Persistence determines the retention memory of the factors responsible for variations in a system, in this case monthly NO_2 and O_3 variation. Figure 4 shows ACF for Nairobi with a time lag of 1 to 12 months. Although in general, one should test for autocorrelation at lags one to lag n/4, where n is the total number of observations in the analysis, estimates at longer lags have been shown to be statistically unreliable (Box and Jenkins, 1976). In some cases, the effect of autocorrelation at smaller lags will influence the estimate of autocorrelation at longer lags resulting in an apparent correlation, even though no direct correlation exists. Although the Partial Autocorrelation Function (PACF) removes the effect of shorter lag autocorrelation from the correlation estimate at longer lags, the estimate is only valid to one decimal place. ACF and PACF each vary between plus and minus one, with values closer to plus or minus one indicating strong correlation. Negative ACF means that a positive return for one observation increases the probability of having a negative return for another observation, depending on the lag and vice-versa. The confidence limits are provided to show when ACF or PACF appears to be significantly different from zero. In other words, lags having values outside these limits (shown as blue dotted lines) should be considered to have significant correlation.

The systems of both NO_2 and O_3 autocorrelations became decoupled after a one-month lag indicated by non-persistence in the autocorrelations as the lag increases. This is attributed to the short memory of pollutant controlling factors. A remarkable feature noticed in the ACFs is the transformation of sign from positive to negative at lag 3. This may be due to the 3-month contrasting wind systems over Nairobi. NO_2 ACF achieved stability between lag 5 and 6 and then changed sign for the second time from negative to positive at lag 10. Ozone ACF achieved stability between lag 8 and 9 and then

changed sign for the second time from negative to positive at lag 11. This stabilization may be attributed to active weather systems during those 2-3 months. Active weather ensures strong mixing and vigorous transport of pollutants. During periods of active weather the incursion/exchange of ozone due to interplay of stratospheric and tropospheric elements is enhanced.

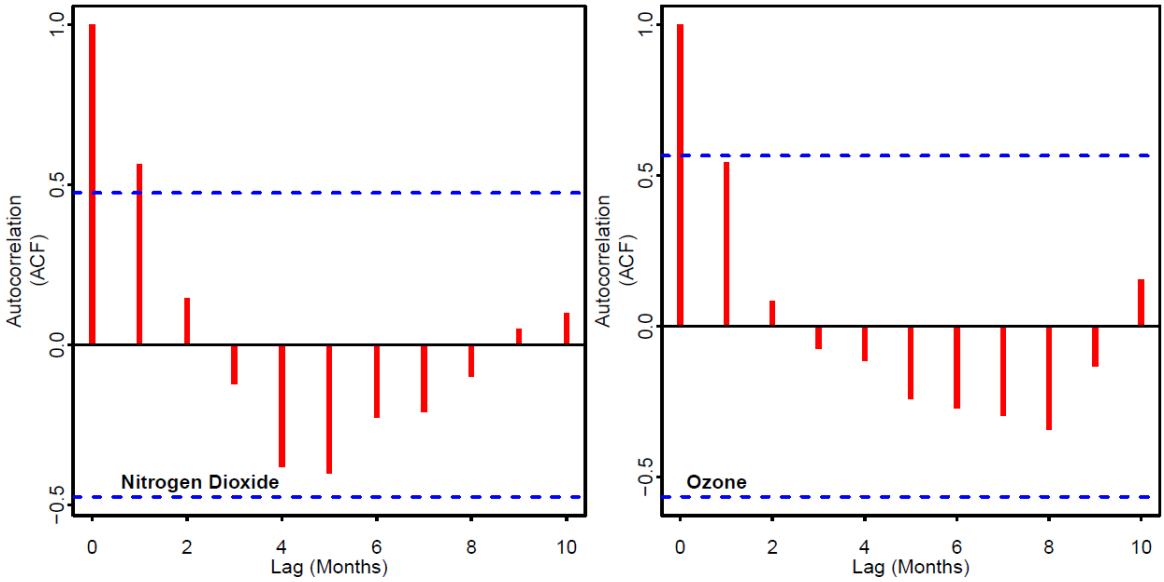

Figure 5: Autocorrelation Functions (ACF) for Nairobi with a time lag of 1 to 12 months

The relationship between monthly NO_2 and O_3 time series was described by calculating the cross correlation functions (CCFs). Generally, in the relationship between two time series (y_t and x_t), the series y_t may be related to past lags of the x-series. In such a case, the sample cross correlation function (CCF) is helpful for identifying lags of the x-variable (NO_2) that might be useful predictors of y_t (O_3). In R, the *sample CCF* is defined as the set of sample correlations between x_{t+k} and y_t for $k = 0, \pm1, \pm2, \pm3$, and so on. A negative value for k is a correlation between the x-variable at a time before t and the y-variable at time t, and x is said to be leading y. When h is *positive* x is said to be lagging y.

Although in some problems, the goal may be to identify which variable is leading and which is lagging, in this case the x-variable (NO_2) was examined to be leading variable of the y-variable (Ozone).

Therefore, since we will want to use values of the x-variable to predict future values of y, report what is happening at the negative values of h on the CCF plot.

The most significant and positive cross correlations occur at k=-1 and 0, indicating that above average concentration of NO_2 is likely to lead to above average levels of O_3 during the same month (r=0.79) or a month later (r=0.7). And, below average concentrations of NO_2 is associated with a likely below average O_3 levels during the same month or a month later. The other relationship shows a dominant negative cross correlations occurring somewhere between k=–6 and about k=–4. This indicates that above average concentration of NO_2 likely to lead to below average levels of O_3 about 5 months later (r=0.39). And, below average concentrations of NO_2 is associated with a likely above average O_3 levels about 5 months later.

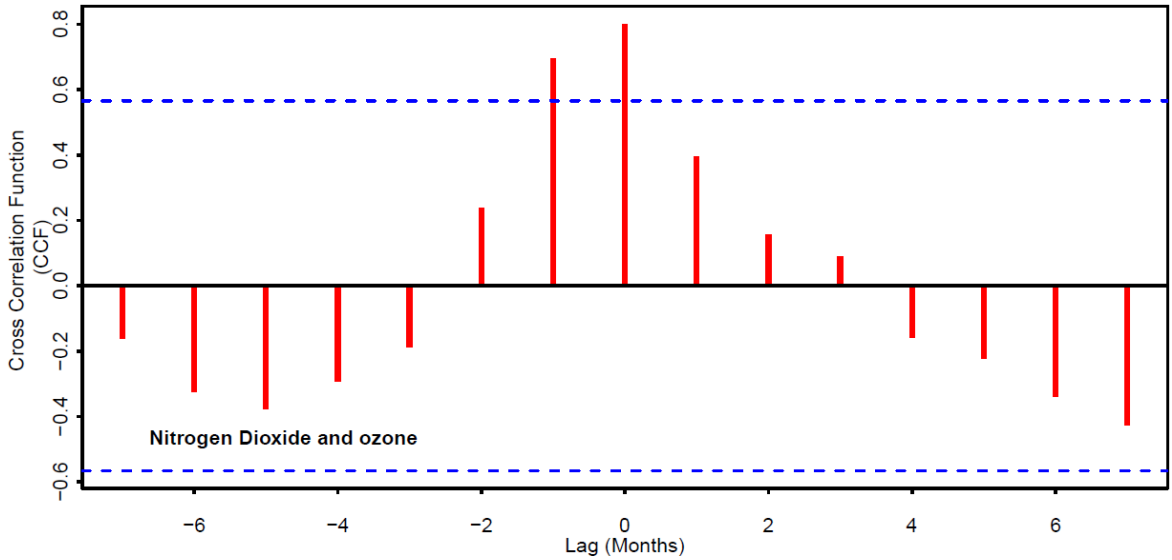

Figure 6: Crosscorrelation Functions (CCF) for Nairobi with a time lag of 1 to 7 months

Interannual Variation

Figure 5 shows interannual variations of average NO_2 and O_3 concentrations for five years. The interannual variation presents an interesting feature in which both NO_2 and O_3 concentrations oscillate biennially. NO_2 dipped in 2010 and peaked in 2012 before dipping again. O_3 increased in 2011 but plunged in 2012 and increased again in 2013. Several studies have termed the observed interannual oscillation of O_3 as a footprint of quasi–biennial oscillation (QBO) that occurs every two years, and that dominates the ozone variability in the tropics (Chehade *et al*., 2014; Logan *et al*., 2003). QBO is a global upper level atmospheric circulation. Its activity has marked effects on wind regimes that control rainfall. Since these wind systems are a factor in ozone variations, QBO activity therefore appears to aid O_3 accumulation because the effect of the circulation that trickles down from upper atmosphere could transport O_3 to the stratosphere and troposphere. Tropospheric O_3 anomalies associated with the QBO of about 10-20% that of typical tropospheric O_3 values (Lee *et al*., 2010). O_3 accumulation due to the QBO effects, comparing years 2009 and 2010 only, could be as high as 2.24% over Nairobi (Lee *et al*., 2010). Effects of ENSO on O_3 have also been observed. Negative tropospheric O_3 anomalies have been reported over Nairobi consistent with anomalous upwelling expected during ENSO events (Lee *et al*., 2010). According to Chehade *et al*. (2014), the signature of the 11-year solar cycle also covers all latitudes and contributes about 10 DU of O_3 from solar maximum to solar minimum.

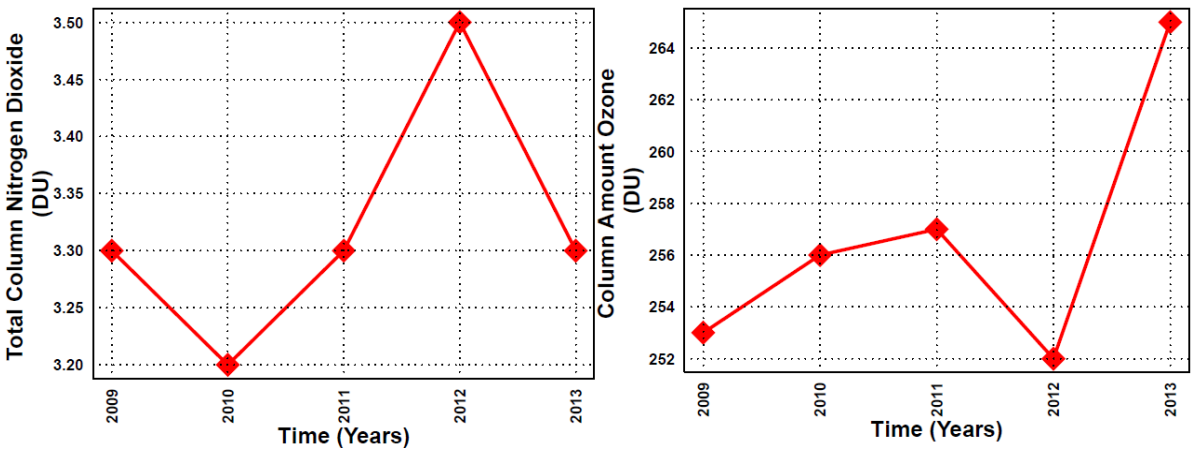

Figure 7: Interannual variations of average NO2 and O3 concentrations for five years

The analysis of the year to year variation revealed increasing trends of both NO_2 and O_3 concentrations over Nairobi. Average increasing rates of 0.01 DU/year (0.27% per year compared to mean value) and 1.19 DU/year (0.46% per year compared to mean value) for NO_2 and O_3 concentrations, respectively. Similarly increasing O_3 trends (0.89DU/yr or 0.35% $year^{-1}$) have been observed over Nairobi by Ogunjobi *et al*. (2007). The increasing rate of ozone could be aggravated in the face of increasing anthropogenic activities. An increase (~10 ppbv) in surface and lower tropospheric ozone in all seasons except in summer is attributed to an increase in urban-industrial emissions in response to the corresponding population increase (Diab *et al*., 2004). An increase in the domestic use of biofuels for heating in poor urban settlements is expected to account for the contrasts between summer and other seasons.

Conclusions

The temporal evolution of NO_2 and O_3 over Nairobi has been analysed. The role of rainfall producing mechanism has been stressed. NO_2 and O_3 distribution is mostly controlled by thunderstorms and extreme rainfall and intrusion of stratospheric air. This is such that their accumulation follows synchronously the migration of wind systems. The relationship between monthly NO_2 and O_3 is such that, above average concentration of NO2 is likely to lead to above average levels of O3 during the same month (r=0.79) and below average levels about 5 months later (r=0.39). As reported by Chehade *et al*. (2014), oscillating biennial feature exhibited by NO_2 and O_3 interannual trend is considered as the footprints of the upper level atmospheric circulation referred to as quasi biennial oscillation (QBO). This circulation acts to increase the exchange of ozone between the stratosphere and the troposphere.

It has also been shown that NO_2 and O_3 are increasing at annual average rates of about 0.27% and 0.46% per year compared to mean values, respectively. The increasing rate of ozone could be aggravated in the face of increasing urban-industrial emissions in response to the corresponding population increase (Diab *et al*., 2004). Daily variation of both NO_2 and O_3 depicts stagnating trends over the entire period of study. This difference is attributed to the fact that, whereas daily NO_2 and O_3 are influenced by mechanisms that control the gradual transition of dry period to wet period within the course of a year, interannual variability is driven by the differences in each year's general weather conditions.

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