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Study of variations in Columnar Ozone Concentration at Rajkot

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ABSTRACT
The ozone data obtained from Total Ozone Mapping Spectrometer (TOMS) and Solar Back scatter Ultra Violet spectrometer (SBUV) on Nimbus 7 satellite have been used to study the variability of the total column amount of ozone and ozone concentration in different atmospheric layers respectively at Rajkot (22.3^{0}N, 70.8 ^{0}E) over a period from 1980 –2004. An examination of the monthly mean values obtained from TOMS indicates a marked seasonal variation with a maximum around June and a minimum around December.

The SBUV profile of springtime ozone indicates that the stratospheric ozone concentration at Rajkot has decreased consistently from 1982 to1999. The concentration is found to have increased suddenly in 2000 and further in 2001. Thereafter it decreased in 2002 and again increased in 2003. An insignificant overall downward trend is observed in the total column amount of ozone (0.85%) from 1997 to 2004. Latitudinal variation of ozone from Srinagar to Kanyakumari has also been studied for the year 2003, which indicates an increase in ozone concentration with latitude. The results are discussed in the light of photochemical and dynamical effects.

INTRODUCTION

Atmosphere is the medium for life on the surface of the planet and is the transition zone between earth and space. It comprises of a mixture of gases exposed to the electromagnetic spectrum of the sun. The atmosphere does not have a well-defined upper boundary but instead its density decreases progressively with altitude and ultimately merges with the space environment. The intermediate region, which extends from 10 to 100 km altitude, is generally called the middle atmosphere. One of the most important constituents in the middle atmosphere is ozone, because it is the only atmospheric species that effectively absorbs ultraviolet solar radiation from 2000 ^{0}A to 3000 ^{0}A, protecting plant and animal life from exposure to harmful radiation. (Brasseur & Solomon1986). Ozone is the key trace constituent that participates in the chemistry and radiation of the earth's middle atmosphere. Hence a decrease in stratospheric ozone will result in an increase in ultraviolet B radiation, which will have negative impacts on human health. However as ozone is toxic to the living system, elevated tropospheric ozone concentration will damage the tissues of plants and animals and will also cause the temperature of the atmosphere to rise. Thus there is a great need to monitor the atmospheric ozone concentration. The recognition in the early seventies of the possibility of a long term global ozone depletion due to catalytic reactions in the stratosphere involving chemicals released by anthropogenic activities and its possible impact on the biosphere as well as the perturbation in the middle atmosphere radiation budget has put ozone on the center stage of atmospheric research over the past few decades. (Subbaraya & Lal 1999) The discovery of the Antarctic ozone hole in the mid eighties by Joseph Farman, Brian Gardiner and Jonathan Shanklin; a research group from The British Antarctic Survey highlighted the gravity of the problem. This was responsible for triggering intense scientific research and signing of International agreements like Montreal Protocol in 1987 to reduce CFC emissions (WMO 1989).

For the past several decades, ozone has been regularly monitored on a global basis by means of Total Ozone Mapping Spectrometer (TOMS) and Solar Back Scatter Ultraviolet Spectrometer (SBUV) carried on Nimbus -7 satellite. These methods require the sun as the background source of radiation and are therefore limited to daytime observations in good weather conditions. In India, ozone is being monitored since last three decades by India Meteorological department, which include

measurements of total ozone with Dobson Spectrometer as well as profiles of ozone from ozonesondes. There have also been a number of major campaigns like INDOEX, which provide insights of seasonal changes in ozone. Mandal, Beig & Mitra (2004), have studied the ozone scenario over India, and have found that although the total ozone has not changed significantly over the past three decades, there has been a considerable reduction in stratospheric ozone and substantial increase in tropospheric ozone. The study of variations in ozone concentration at Rajkot has special significance because it is located in the tropical region, where most of the ozone formed is due to the availability of the high dose of solar UV radiation. Unlike the observations recorded at Rajkot, Satsangi et al. (2004) have observed higher ozone mixing ratios at Agra during winter, and not in summer.

DATA AND ANALYSIS

The total column amount of ozone at Rajkot and its distribution in various atmospheric layers have been studied using TOMS and SBUV data respectively over the period from 1980 to 2004.TOMS is a source of high-resolution global information about the total ozone content of the atmosphere. It measures the ultraviolet sunlight back scattered from the clouds or the ground to measure the total ozone amount. It cannot measure nighttime ozone or make measurements in the winter polar darkness. The largest amount of satellite based ozone measurements comes from the Solar Back Scattered Ultra Violet Technique (SBUV). In this method, the ratio of sunlight scattered back to the spacecraft from the earth-atmosphere system to that incident at the top of the atmosphere is used to determine the ozone amount. It divides the earth's atmosphere into 13 layers and determines the columnar ozone for each of these layers. The classification of these layers and their corresponding altitude from the surface of the earth is shown in Table 1. This technique works only during daytime and can be used to measure total ozone columns and vertical distribution.

A hot summer and dryness in non- rainy seasons characterize the climate of Rajkot district. The year may be divided into five seasons. The winter season extends from December to February followed by spring in March, summer from April to June, monsoon from July to September and autumn in October and November.

Table 1.

LAYER NUMBER	ALTITUDE in km
1	0.00 to 15.5
2	15.5 to 21.5
3	21.5 to 25.5
4	25.5 to 28.5
5	28.5 to 31.5
6	31.5 to 34.5
7	34.5 to 37.5
8	37.5 to 41.5
9	41.5 to 45.0
10	45.0 to 48.0
11	48.0 to 52.5
12	52.5 to 56.5
13	56.5 km to the top of the atmosphere.

RESULTS AND DISCUSSION

Seasonal variation

The monthly mean Total Columnar Ozone values obtained for the location of Rajkot from the TOMS data are plotted against months for the years Aug.1996 to June 2004. (Fig.1). It shows a clear seasonal variation that is repeated consistently year after year. The ozone concentration is found to be maximum around May –June. It decreases during monsoon and autumn and reaches a minimum in December. Thereafter it gradually increases throughout the winter and spring and reaches a peak in summer. This trend is consistently observed in all the years from 1996 to 2004 (as shown in Fig.1). Chakrabarty & Chakrabarty (1979) have observed similar results for other Indian stations.

The amount of ozone in the upper stratosphere (above 40kms) is about 3% of the total ozone. Thus the variations occurring above 40 kms have little measurable effect on the total ozone column. The variation of ozone in the lower Stratosphere where its density is maximum (87%) and in the Troposphere (10%) has a measurable effect on the total ozone column. The amount of ozone in the Troposphere and lower Stratosphere in general depends on both dynamics and chemistry. The dynamical influences include wave driving of the stratospheric circulation and Tropopause folds. The chemistry part includes the photochemical production and destruction of O_3.

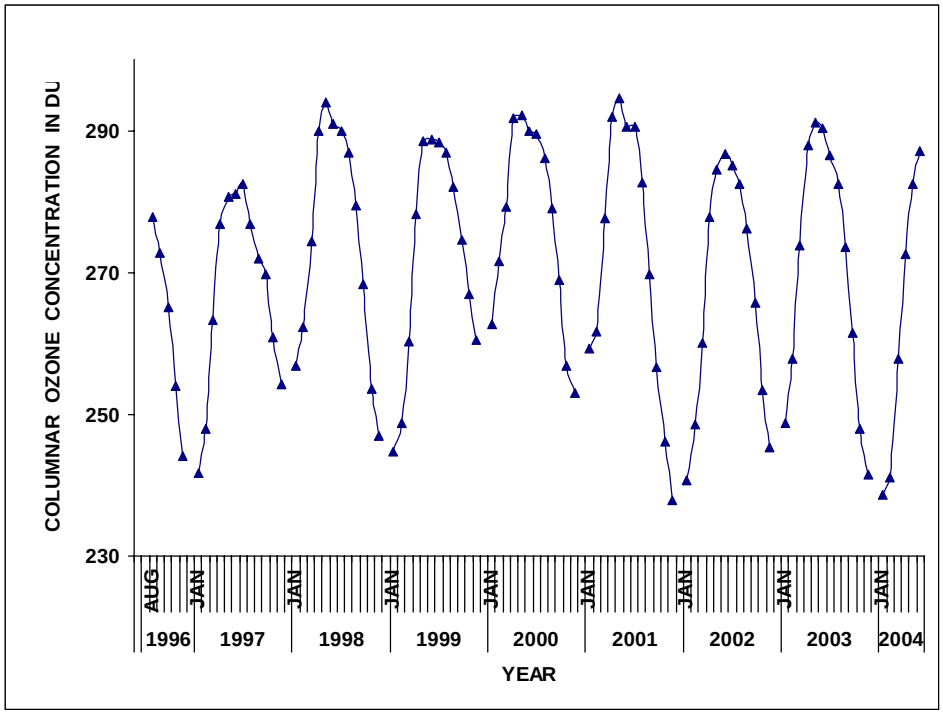

Figure 1: Total columnar ozone measured by TOMS over Rajkot from August 1996 to June 2004.

Seasonal variation of total columnar ozone is difficult to account for, since the major contribution to total columnar ozone comes from the lower Stratosphere, where photochemical life times are of the order of several months to years. (Banks & Kockarts; 1973) Thus short-term changes in columnar ozone are not expected unless there is a significant variation in Tropospheric ozone below 5 kms.

The primary cause of large values of ozone in June may be the high solar flux acting upon a pool of accumulated NOx and hydrocarbons built up during winter resulting in local photochemical production. (Vingarzan Roxanne 2004)

$OH + CO \rightarrow H + CO_2$

$H + O_2 + M \rightarrow HO_2 + M$

$HO_2 + NO \rightarrow NO_2 + OH$

$NO_2 + \text{sunlight} \rightarrow NO + O$

$O + O_2 + M \rightarrow O_3 + M$

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

The reaction rate coefficients that determine photochemical loss rates are temperature dependant. The rate coefficient for the reaction $O+O_3 \rightarrow 2\ O_2$ as a function of temperature is given by: $K\ (O, O_3) = 8\ X10^{-12}\ e^{-2060/T}$cc/mol.sec

Thus the loss rate of ozone decreases by 30% for every 10 deg fall in temperature. The tropical region is also a source of Kelvin waves (Hirota 1979). It is now realized that, apart from advection, diffusion transport due to waves over a large range of temporal and spatial scale sizes, plays an important role in the net transport and there is a close coupling of chemistry and dynamics (Subbaraya et al.1994)

Another factor that contributes for the increase in ozone concentration is long photochemical lifetime of ozone in winter (approximately 200 days) in the lower stratosphere. This is because the sun's angle is quite low during winter and the lower stratospheric ozone is effectively shielded from the UV radiation by the ozone in the upper stratosphere. Thus ozone in the lower stratosphere is safe from destruction by photochemical processes. This accumulation of ozone rich air over the entire winter period leads to an increase in concentration in winter.

A factor more recently linked to the annual ozone cycle is the inter continental transport of pollution. The same atmospheric mechanism responsible for the transport of Asian desert dust during spring has been shown to transport primary emissions and ozone. (Vingarzan Roxanne 2004).

The prominent wind directions during summer are N, NW and W. During monsoon, winds blow from the SW direction. During winter NE, NW and N flow is predominant (Sahu 2004; Chand, Lal & Naja 2003)

The general wind pattern from NW & N direction takes the polluted continental air down to Rajkot during summer. The Asian region is a fast developing

region with increasing levels of pollution from industries and other manmade sources. O_3 is produced by the photo oxidation of pollutants (carbon monoxide & hydrocarbons) in the presence of adequate amount of nitrogen oxides at lower altitudes. Once O_3 is produced it gets transported along with the wind resulting in a high concentration in summer.

$CH_4 + OH \rightarrow CH_3 + H_2O$
$CH_3 + O_2 + M \rightarrow CH_3O_2 + M$
$CH_3O_2 + NO \rightarrow CH_3O + NO_2$
$NO_2 + hn \rightarrow NO + O$
$O + O_2 + M \rightarrow O_3 + M$
$CH_3O + O_2 \rightarrow CH_2O + HO_2$
$HO_2 + NO \rightarrow NO_2 + OH$
$NO_2 + hn \rightarrow NO + O$
$O + O_2 + M \rightarrow O_3 + M$
$CH_2O + hn \rightarrow CO + H_2$
$CO + OH \rightarrow CO_2 + H$
$H + O_2 + M \rightarrow HO_2 + M$
$HO_2 + NO \rightarrow OH + NO_2$
$NO_2 + hn \rightarrow NO + O$
$O + O_2 + M \rightarrow O_3 + M$
Net; $CH_4 + 6O_2 + hn \rightarrow H_2O + H_2 + 3O_3 + CO_2$

The Arabian Sea lies to the south and west of Rajkot. During monsoon the pristine air from the Arabian Sea blows over Rajkot resulting in decrease in O_3 concentration due to wash out effects by pure monsoonal air masses.

Solar cycle variation

Variation of ozone trends with sunspot activity has been studied for Rajkot from 1980-2003 (Figs 2 & 3.) The output of solar UV radiation was influenced by magnetically active regions (sun spots) on the sun. The period when sunspot activity is at its greatest during the 11-year cycle is solar maximum (eg.1989 and 2000), while the period when sunspot activity is at it's least is a solar minimum (eg.1986 and 1996). The UV output of the sun increases as sunspot activity increases.

Table 2. Approximate variability of solar UV radiation over solar cycle as a function of wavelength

Wavelengh	Variation
2000 Å	8%
2200 Å	4%
2700 Å	1%
3000 Å	0.2%

This shows that the production of ozone should be more sensitive to variations in solar UV output (4 to 8%) than the loss rate of ozone. Thus the production rate of ozone should be high when solar activity is at its maximum and decrease when solar activity is at

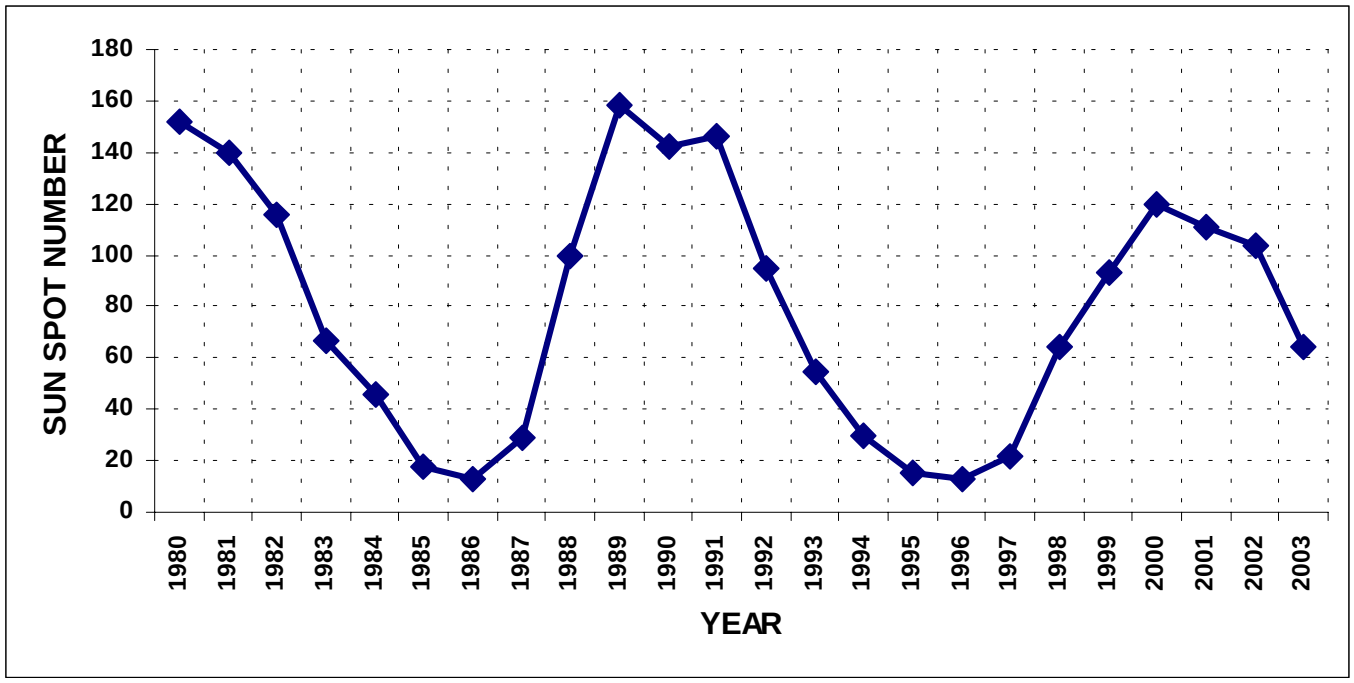

Figure 2. 11 Year solar cycle.

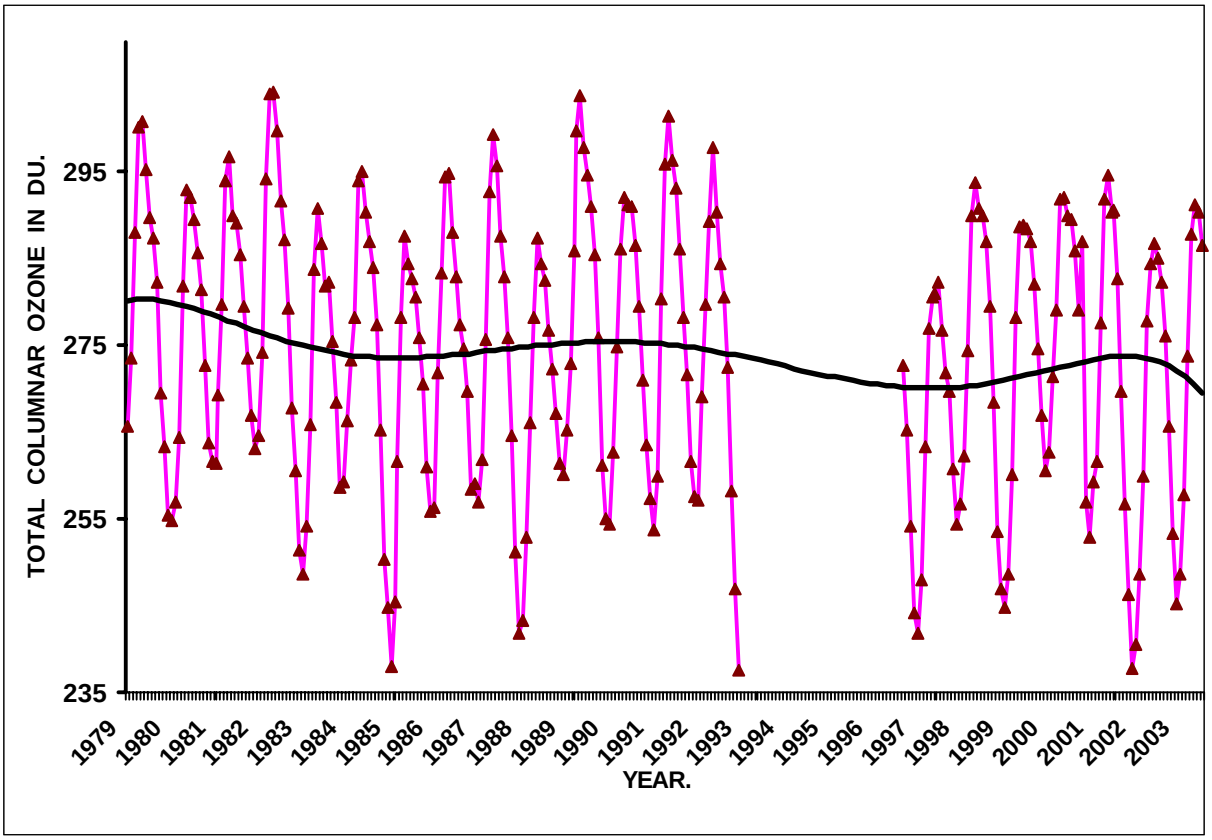

Figure 3. Columnar ozone measured by TOMS over Rajkot from 1979 to 2003.

its minimum. It is found that the monthly averaged ozone variation is in phase with the monthly averaged sunspot variations. Ruderman & Chamberlain (1975) have suggested that during solar minimum condition, there is an increase in the low energy cosmic rays in the earth's atmosphere, which will increase NO_x and decrease ozone. Thus the ozone concentration is expected to decrease during solar minima, which is observed in this study (as in 1986 and 1996). Similarly, the ozone concentration during solar maxima (1989 and 2000) is found to be higher than that observed during the other remaining years of the solar cycle.

El Nino Southern Oscillation

One of the most intense EL Nino episodes in more than a century occurred during 1997-1998.This strong EL Nino might have influenced the total ozone column over the Indian subcontinent, leading to an increase in total ozone column in 1997-1998. Singh, Sarkar and Singh, 2002; investigated the effect of sea surface temperature on the total ozone column and found a close relation with the increasing ozone trend during the EL Nino period.

Latitudinal variation

The monthly mean ozone data for different Indian stations obtained from TOMS indicates that the columnar ozone concentration increases as we move northwards from Kanyakumari to Srinagar (Fig.4) i.e. with increasing latitude. This is because ozone is produced at the tropics and then transported to the poles where it is relatively inert photo chemically. Chakrabarty & Chakrabarty;(1979) have observed similar results for other Indian stations

The Hadley cell circulation observed in India is different compared to that observed globally due to the presence of the Himalayas (Chakrabarty, Chakrabarty and Beig 1984; Mani & Sreedharan, 1973; Chakrabarty & Peshin 1997) The air ascends near 24^0N and descends in the higher latitudes near the Himalayas to give a pole ward drift in the upper air, with a return current towards $24\ ^0$N at the surface. The upwelling of the Hadley cell circulation is usually felt up to the tropopause, but sometimes it also reaches the lower stratosphere. This reverse Hadley cell circulation may have a pronounced effect on the distribution of the ozone at different locations in the Indian subcontinent

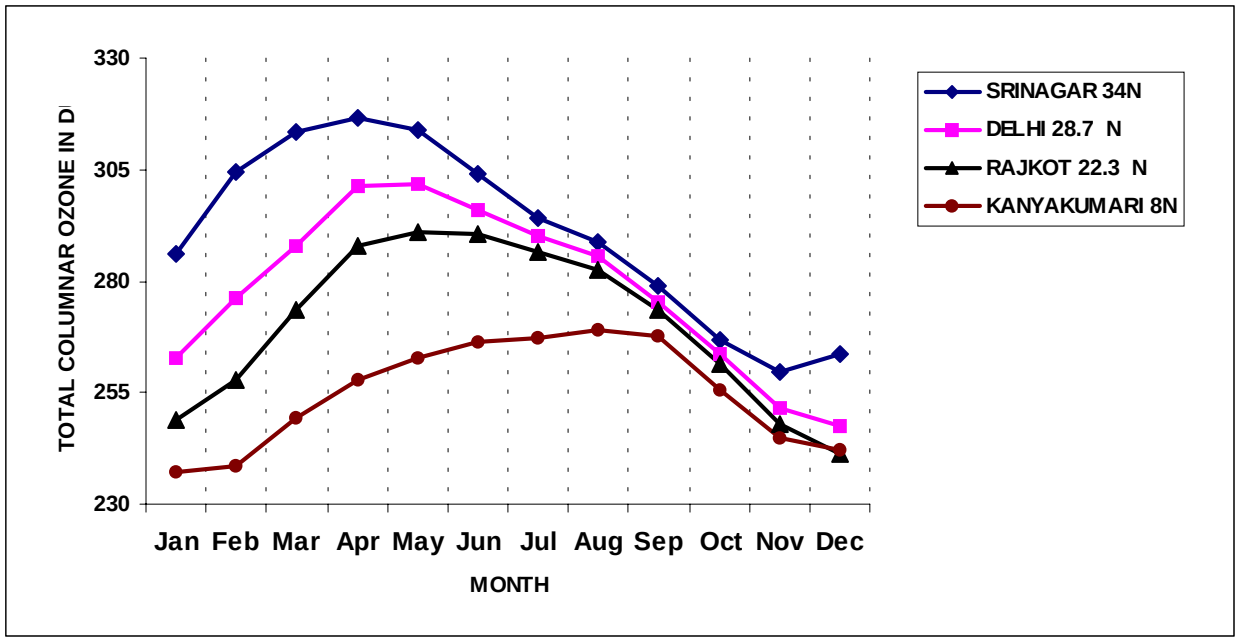

Figure 4: Latitudinal variation of total columnar ozone measured by TOMS in the year 2003.

resulting in an earlier peak ozone concentration at Srinagar (34^{0}N) compared to Kanyakumari (8^{0}N). The atmospheric circulation is such that it transports ozone during winter from low latitudes to poles. This transport is responsible for springtime maxima at higher latitudes. The transport is less rapid during summer, and the maxima gradually decays and moves to lower latitudes. (Goody & Walker 1972.)

Trend analysis

Singh, Sarkar & Singh (2002) have reported an increasing trend in columnar ozone for the period 1996-2000 for Kodaikanal and Delhi, which matches with the observations for Rajkot (Fig.3). However if the period of observation is extended to 2004, then an overall downward trend in the total columnar O_3 is observed from 1996 to 2004 (Fig.1). It is observed from the trend analysis, that there is an insignificant change in the columnar ozone concentration from 1996 to 2004, which may be due to the global decrease in CO concentration. (Novelli. et al.1994; Khalil and Rasmussen 1994). The production of tropospheric ozone is mainly from CO and CH_4 in presence of hydrocarbons and nitrogen oxides. Hence a decrease in chemical fuels may ultimately lead to a decrease in production of NO_2, consequently affecting the ozone production. (Saraf & Beig 2004.)

The average ozone concentration from 1st to 15th March has been used to study the distribution of ozone and its variations in different atmospheric layers during springtime at Rajkot using SBUV data. It is observed that the stratospheric ozone concentration at Rajkot has decreased consistently from 1982 to 1999. (Fig. 5) This may be due to the catalytic destruction of ozone by compounds containing Bromine, chlorine, fluorine, carbon (chloro fluro carbons) and oxides of Nitrogen (NO_x), which are produced in the stratosphere by the reaction of O ^{1}D with N_2O (which is released from the biosphere below) and from the emission of supersonic aircraft. NO_x is the most important destroyer of ozone in the 25-45 Kms altitude region. Br takes part in catalytic destruction of ozone and is 40 times more reactive than CL_2 in ozone depletion

The catalytic cycles of ozone destruction can be illustrated by

$O_3 + X \rightarrow O_2 + OX$

$OX + O \rightarrow O_2 + X$

NET: $O_3 + O \rightarrow 2O_2$

where X is a radical, which goes through the catalytic cycle, destroys ozone and is recovered at the end of the reaction cycle. X can belong to the NO_X, CLO_X, HO_X or Br family. (Subbaraya & d Lal 1999).

The ozone concentration is found to have increased suddenly in 2000 and further in 2001. Thereafter it decreased in 2002 and again increased in 2003. This recovery may be attributed to the international agreements like Montreal Protocol to reduce emissions due to CFC'S.

It is observed from Fig 5, that the ozone concentration is different for different years up to an

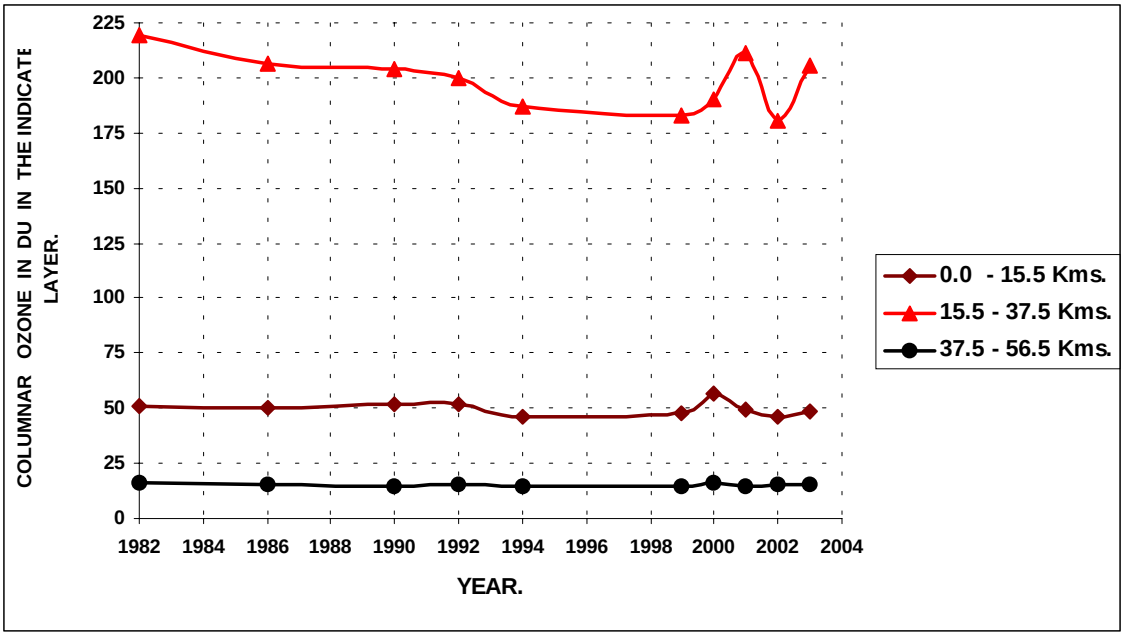

Figure 5: Springtime variation of ozone in different atmospheric layers from 1982 to 2003 at Rajkot.

altitude of approximately 37.5 kms. Beyond 37.5 kms the O_3 profile is found to remain constant from 1982-2003. Maximum variation in ozone concentration is observed in the altitude range from 15.5 to 37.5 kms. Thus the variation in ozone concentration in this altitude range has a measurable effect on the total column amount of ozone.

CONCLUSIONS

The monthly mean values of ozone obtained from TOMS indicate a marked seasonal variation with a maximum around June and a minimum around December .A study of springtime ozone in different atmospheric layers from 1982-2003 indicates that the stratospheric ozone concentration at Rajkot has decreased consistently from 1982 to 1999. The concentration is found to have increased suddenly in 2000 and further in 2001. Thereafter it decreased in 2002 and again increased in 2003.The ozone concentration in India is found to peak earlier at places located at higher latitudes during summer compared to places at lower latitudes.

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