VERTICAL DISTRIBUTION OF OZONE AT PRETORIA: COMPARISONS BETWEEN 1965-68 AND 1990-91

Mark Zunckel[1], Roseanne D. Diab[2] and Malcolm W.J. Scourfield[3]

1. South African Weather Bureau, Private Bag X97, Pretoria, South Africa, 0001
2. Department of Geographical and Environmental Sciences, University of Natal, King George V Avenue, Durban, South Africa, 4001
3. Space Physics Research Institute, Department of Physics, University of Natal, King George V Avenue, Durban, South Africa, 4001

INTRODUCTION

Ozone in the stratosphere absorbs most of the ultraviolet radiation of wavelengths shorter than 320 nm emitted from the sun. Increased UV radiation of these short wavelengths reaching the earth's surface, through decreases in stratospheric ozone, can have negative effects on humans[1] and much of the biosphere[2,3]. Decreases in total ozone were first reported by Farman *et al*[4], from ground based Dobson spectrophotometer measurements at Halley Bay, and were confirmed using Total Ozone Mapping Spectrometer (TOMS) satellite data.[5] These decreases were later shown to be global in extent[6].

While stratospheric ozone concentrations have decreased, tropospheric ozone concentrations over Europe and the central and eastern United States were shown to have increased by 20% to 100% since 1940.[7] Using data from the Montsouris Observatory, increases in tropospheric ozone concentration were found in the Great Lakes area of the United States and in Europe since the middle on the 19th century[8,9]. Ozone is a strongly corrosive pollutant, having detrimental effects on vegetation[10] and on human health[11]. Increasing concentrations of tropospheric ozone are therefore cause for concern.

Ozone was monitored above Pretoria, South Africa, during the 1960's and early 1970's, measuring total ozone and the Umkehr effect[12], from which the vertical distribution of ozone was determined. The main emphasis focused on trace gas movement and related stratospheric flow. With growing international awareness of the destruction of the ozone layer, the South African Weather Bureau reactivated its ozone monitoring program in 1989 and the South African National Ozone Research Program (SANORP), was launched shortly afterwards.

Ozone monitoring during the earlier period was carried out before the main onset of global ozone depletion and before explosive industrialization in the areas surrounding Pretoria. These data, henceforth termed 'background data', provide a useful background against which current values may be compared. In order to evaluate changes in ozone concentration since the earlier monitoring period a benchmark ozone climatology was established in studies of the seasonal cycle of total ozone over Pretoria, 25°44'S and 28°11'E, from 1964 to 1972[13] and the vertical distribution of ozone above Pretoria from 1965 to 1968[14]. The latter study showed that the mean ozone concentration increased rapidly with height in all months from partial pressure values of less than 1 mPa at the surface to about 14 mPa between 24 and 30 km. The concentration then steadily decreased to values below 1 mPa above 54 km.

Being positioned relatively close to the area of the most drastic form of ozone depletion, the seasonal Antarctic ozone hole, it is of fundamental importance to monitor changes in ozone concentration above South Africa and to understand the mechanisms that control these changes. These are research topics being addressed by SANORP. As an initial step, this paper examines mean monthly total ozone above Pretoria from August 1989 to May 1990, and from July 1990 to September 1991 above Irene, situated approximately 20 km south of Pretoria at 25°15'S and 28°22'E. It also examines vertical ozone profiles derived from the Umkehr effect for the period July 1990 to June 1991, at Irene. Comparisons of these recent data are made with the background data. Suggested hypotheses are made in an attempt to explain the changes that have occurred.

OBSERVATIONAL DATA

The ozone data used in this paper were obtained from spectroscopic measurements of total ozone and of the Umkehr effect, and from electrochemical cell (ECC) ozonesonde soundings. Mean daily values of total ozone were obtained from at least three measurements taken during the day, using Dobson spectrophotometer No. 89, monitoring A and D wavelength pairs. These daily means were averaged to give a mean total ozone value for each month. Data were collected in Pretoria, 25°44'S and 28°11'E, during the background period from August 1989 to May 1990, but the monitoring site was relocated to Irene, approximately 20 km south of Pretoria at 25°15'S and 28°22'E, in June 1990. Due to the close proximity of the two monitoring sites, the data set is assumed to be continuous.

Measurements of the Umkehr effect were done less frequently. The vertical distribution of ozone was calculated from these measurements[12] and these were used to derive mean monthly ozone profiles. Data from weekly ECC ozonesonde sounding were used to examine tropospheric ozone in more detail. The number of each observation type used to determine the vertical distribution of ozone is listed in Table 1. Ozone concentrations measured by the sondes were normalized to a simultaneous ground based measurement of total ozone. The ozone data presented in this paper were submitted to the World Ozone Data Centre for archiving.

Table 1: Number of observations of the vertical distribution of ozone using the Umkehr method and using balloon-borne ECC ozonesondes, for 1965-68 and 1989-91.

	Number of umkehrs per year						Sondes	
Month	1965	1966	1967	1968	1990	1991	1990	1991
1		19	9	13		3		3
2		17	8	7		7		4
3		30	14	7		3		4
4		20	6	6		12		4
5	20	25		10		7		5
6	25	27	22	11		6		4
7	22	25	26	11	10	10	1	4
8	21	23	12	12	6	9	1	4
9	16	18	13	14	9	4	2	3
10	19	18	10	6	5	7	4	5
11	15	9	9	8	5		4	
12	18	9	9	6	2		4	
Total	156	240	138	111	37	68	16	40

COMPARISON BETWEEN BACKGROUND AND RECENT DATA

In Figure 1 the mean monthly total ozone data from the background period is compared with corresponding data from the recent period. The lower graph represents the mean seasonal cycle for the period 1964 to 1972[13], showing mean monthly values ranging between 245 DU and 277 DU, about an annual mean of 259 DU. Mean total ozone values from August 1989 to May 1990 at Pretoria, and from July 1990 to September 1991 at Irene were used to calculate the monthly means indicated by the upper graph in Figure 1. The seasonal cycle is consistent between the earlier and the more recent data periods, with mean monthly values ranging between 316 DU and 273 DU. However, there is currently more ozone in the atmospheric column above Pretoria and surrounding areas than during the background period. The annual mean of the seasonal total ozone cycle for the recent period is 289 DU, representing an increase of 12% over the background mean.

Comparisons of the vertical distribution of ozone are made with reference to the 9 standard Umkehr layers[12], with layer 1 extending from the surface, at 1523 m above sea level (asl), up to 6 km, then layers 2 to 9 at 6 km intervals up to 54 km. The mean monthly background data and the corresponding data for the 12 months July 1990 to June 1991 in each of the 9 Umkehr layers is shown in Figure 2. There are three points which are worthy of note. Firstly, in layers 1 and 2, the tropospheric layers, a significant increase in ozone concentration relative to the background is shown; secondly, the third to seventh layers, covering the tropopause and the stratosphere up to 42 km and including the ozone layer, show a relative decrease in ozone concentration; and thirdly, layers 8 and 9, comprising the stratosphere above 42 km, show little change relative to the background data. These changes in ozone concentration in the 9 Umkehr layers are summarized in Table 2.

Table 2: Algebraic difference in ozone concentration (mPa) in the 9 standard Umkehr layers between the background and current data. The mean concentration (mPa) in each layer for both periods is listed and the mean percentage increases.

Month	Umkehr Layer								
	1	2	3	4	5	6	7	8	9
Jan	2.4	1.4	-0.1	-1.1	-1.2	-1.1	-10	0.3	0.3
Feb	2.9	0.8	-1.3	-1.4	-0.7	-0.3	0	0.2	0.2
March	2.6	0.9	-1.2	-1.2	-0.4	-0.1	-3	0.0	0.1
April	2.1	0.9	-0.6	-0.6	-0.1	0.1	-1	0.0	0.1
May	2.1	1.0	-0.8	-0.9	-0.6	-0.2	-9	0.0	0.0
June	1.7	0.9	0.1	0.3	0.2	0.4	2	-0.1	-0.1
July	2.4	0.9	-1.5	-0.9	-0.4	-0.2	-5	-0.2	-1.1
Aug	1.8	1.2	0.2	-0.5	-0.7	-0.8	-11	-0.1	0.0
Sept	3.2	0.7	-1.9	-1.3	-0.9	-0.1	-4	-0.3	-0.1
Oct	2.5	1.1	-1.0	-1.2	-1.5	-0.5	-3	0.0	0.1
Nov	2.0	0.9	-1.1	-1.1	-0.7	-0.1	-1	-0.2	-0.2
Dec	2.4	1.5	0.9	-0.5	-1.2	-1.5	-12	0.1	0.1
Mean 1965-68	0.4	2.1	4.8	10.3	14.1	9.4	4.7	1.9	0.55
Mean 1989-91	2.7	3.2	4.1	9.4	13.4	9.1	4.4	1.9	0.59
Mean % increase	575	52	-15	-9	-5	-3	-6	-	-

The changes in tropospheric ozone and ozone in the region of the tropopause and the ozone layer will be elaborated upon with reference to the following assumed atmospheric divisions: ozone in the lowest two layers, from the surface to 12 km, is purely tropospheric ozone; the third layer usually includes the tropopause, so the ozone in this layer contains both tropospheric and stratospheric ozone; ozone in layers 4 to 6, between 18 km and 36 km comprises more than 65% of the total ozone[13] and includes the 'ozone layer'. Ozone above 36 km is assumed to be middle and upper stratospheric ozone.

TROPOSPHERIC OZONE

Concentrations of tropospheric ozone in the lowest 6 km asl above Pretoria have increased from mean monthly partial pressure values of less than 1 mPa during the background period, to corresponding means of between 2 and 4 mPa during the period July 1990 to June 1991. In the tropospheric layer extending from 6 to 12 km asl, mean monthly ozone concentrations have increased from values of between 1 and 3 mPa during the earlier period, to corresponding mean values of between 2 and 4 mPa during the later period. These statistics represent a mean annual increase of more than four fold in the lowest 6 km asl and a increase of 55% in the layer between 6 and 12 km asl.

It has been well documented[15-18] that nitrogen oxides (NOx), hydrocarbons and carbon monoxide (CO), released during combustion of fossil fuels, react photochemically in sunlight to form ozone. Common sources of these ozone precursors are motor vehicle exhausts, coal burning, industry and biomass burning. Motor vehicles, due to a high Nox/hydrocarbon ratio are potentially the largest contributor to photochemically produced ozone[19].

It is hypothesised that the increase in tropospheric ozone found in the Pretoria area, shown in layers 1 and 2 in Figure 2, is due largely to an increase in the number of NOx, hydrocarbon and CO sources over the last two decades in Pretoria and surrounding areas. Due to the potentially long life of photochemically produced ozone[20], sources further afield could also play a role in these tropospheric ozone increases. For this reason, motor vehicle numbers are considered both locally and nationally and electricity production by coal burning, mostly at sources distant from Pretoria, is also considered. In the absence of NOx data, growth statistics of these two major sources of NOx have been used as proxy data. Due to urbanization and an increase of areas under agriculture, the sources of ozone precursors from natural biomass fires are likely to have decreased[21] and these are not considered as NOx sources contributing to the increase in tropospheric ozone above Pretoria in this paper.

Population growth implies an increase in the number of motor vehicles and in electricity production. Population statistics since 1951 for Pretoria and nationally are shown in Table 3. To satisfy the growing population, demands on motor vehicles and electricity production have also increased. Pretoria and national vehicle number statistics are shown in Figure 3a with commercial vehicles, generally having larger engines which emit more NOx than conventional engines, shown separately. Electricity generation takes place mostly in large coal-fired power stations, by far the greater number of which are concentrated within a 200 km arc to the east and southeast of Pretoria. The growth of electricity production since 1965 is shown in Figure 3b.

Table 3: Population statistics for Pretoria and South Africa for the indicated years (South African Statistics 1988 and Bulletin of Statistics, Quarter ended March 1991, Central Statistics Services)

Year	Pretoria (thousands)	South Africa (millions)
1951	280	13
1960	410	16
1970	543	22
1975	610	
1980	685	25
1985	771	28
1990	869	

It is felt that the growth of NOx sources since the 1960's, shown by Figures 3a and 3b, clearly implies that NOx concentrations have increased over the same period, implying in turn, an increase in the concentration of photochemically produced ozone in the troposphere surrounding Pretoria.

Short episodic occurrences of unusually high ozone concentration in the lowest layer of the atmosphere, from the surface to 6 km asl, have been observed substantiating these claims. An example of two episodes is illustrated in Figure 4. A sequence of observations in two case studies is shown. On day 1 of the first case the ozone concentration in the lowest Umkehr layer is 2 mPa, which is below the mean of 2.67 mPa, shown in Table 2 and Figure 2, for the current period. On the second day, the concentration has more than doubled to 5.3 mPa. On the last day, the concentration has decreased to 1.7 mPa, again below the mean. Note that the second Umkehr layer, 6 to 12 km asl, shows little variation during the episode. A similar sequence of events occurs during the second case, although the episode is of longer duration.

Factors such as the general synoptic flow, turbulence, inversion height, various boundary layer parameters and the location of NOx sources relative to the prevailing winds play a role in the horizontal distribution of tropospheric ozone. Initial investigation has concentrated on four case studies looking at horizontal flow, stability and temperature profiles. In 3 of the 4 cases the peak has occurred with northeasterly winds at 850 hPa, as would be expected if the source of ozone precursors was from the urban plume from the Pretoria CBD, situated to the north of the monitoring site, or from coal burning east of Pretoria. However, in the fourth case the low-level wind was from the southwest. Whether this was due to "recirculated" pollution is not clear at this stage.

Figure 5 shows the mean ozone and temperature profiles for the troposphere at 16 selected pressure levels below 100 hPa. Ozone concentration (mPa) and temperature (°C) were extracted from 49 ozonesonde soundings, released between July 1990 and September 1991, to obtain the mean profiles. Ozone concentration increases with height from a mean value of about 1.7 mPa at the surface, to above 3 mPa at 3175 m asl (700 hPa), or roughly 1600 m above the surface. Above this level, the concentration decreases again to about 2 mPa in the region of the tropopause. It appears as if the bulk of tropospheric ozone is trapped in a layer close to the surface. The depth of this layer corresponds well with the depth of the boundary layer[22] and with the height of the low level jet[23] over the Highveld east of Pretoria. The standard deviation bars on the ozone profile indicate that the largest variability in ozone concentration occurs closest to the ground. This is indicative a relationship between the prevailing weather conditions and the position of the source of ozone precursors relative to the monitoring site. This relationship will be examined in future work.

STRATOSPHERIC OZONE

Ozone concentration in the layer above Pretoria that usually contains the tropopause, 12 to 18 km asl, has decreased from a mean monthly value of 4.8 mPa during the background period, to a corresponding value of 4.1 mPa during the more recent period. In the three layers comprising the ozone layer, from 18 to 36 km asl, the average ozone concentration has decreased from 11.3 mPa in the earlier period to a corresponding mean of 10.6 mPa during the later period. In the middle stratosphere, between 36 and 42 km asl, the mean ozone concentration has decreased from 4.7 mPa in 1965 to 1968 to 4.4 mPa in the period July 1990 to June 1991. Only slight relative changes in ozone concentration, both positive and negative, occurred in the layers above 42 km asl. These statistics represent a relative decrease of 7% in the upper

troposphere and in the stratosphere below 42 km asl, relative to the background period.

Heterogeneous reactions involving chlorine chemistry may account for much of the depletion of ozone with the Antarctic vortex[24,25], but the reasons for these reactions occurring at lower latitudes, accounting for a global scale depletion[6] are less obvious. A possible explanation is a dilution effect[26] where ozone at lower latitudes is reduced through transport of, and mixing with, ozone-poorer Antartic air masses after the breakup of the polar vortex in spring. Analyses of 100 hPa ECMWF wind fields[27] indicate a rapid break up of the polar vortex in October and the re-establishment of meridional flow into and out of the Antarctic stratosphere. This meridional flow could provide the transport mechanism necessary for mixing of ozone-richer air from lower latitudes with ozone-depleted air from Antarctic latitudes after the breakup of the winter vortex. The poleward flow of ozone in the stratosphere above South Africa and the rest of the mid-latitudes cloud result in the observed decreases, contributing to global scale decreases.

Although the current Pretoria data base is limited and the reasons for global depletion of stratospheric ozone are still unclear, it is suggested that the ozone decreases in the region of the tropopause and in the stratosphere above Pretoria, shown in Figure 2 for July 1990 to June 1991, are confirmation of the global decreases in the mid-latitudes of the Southern Hemisphere. A better understanding of the local characteristics of stratospheric ozone is therefore essential in investigating this suggestion.

CONCLUSIOSN

The available data indicate that there has been an increase in total ozone over the last 23 years in the Pretoria area. This is contributed to largely by a significant increase in tropospheric ozone concentrations. In the layer up to 6 km above sea level the average increase was more than 4-fold, while in the next 6 km layer the average increase was 55%. It is hypothesised that this increase is due to increased anthropogenic inputs of ozone precursors such as NOx and hydrocarbons.

Studies of tropospheric ozone in the Northern Hemisphere found substantial increases in concentration since the middle of the 19th century. Although monitoring at Pretoria does not span the same time periods, the increase in low level tropospheric ozone agree with the Northern Hemisphere findings. It is felt that these studies are all indicative of a general global increase of low level ozone, concentrated in industrialized countries.

A relative depletion of 7% in stratospheric ozone and in ozone near the region of the tropopause occurred over the last two decades above Pretoria. Although the data set is limited, it is felt that this decrease is confirmation of the phenomenon of global ozone depletion in the mid-latitudes of the Southern Hemisphere. The mechanisms controlling stratospheric ozone depletion at low latitudes are not well understood, stressing the importance of ozone monitoring and related research in South Africa.

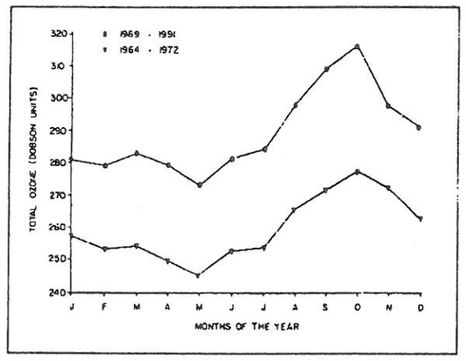

Figure 1: Mean monthly total ozone above Pretoria for 1964-72 compared with monthly means from August 1989 to October 1991.

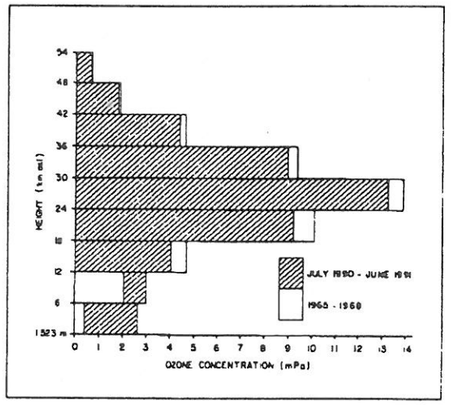

Figure 2: Annual mean ozone concentrating in the 9 Umkehr layers for the period 1965 to 1968 compared with corresponding data for the period July 1990 to June 1991.

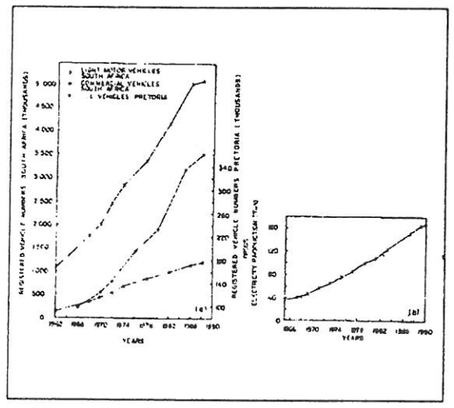

Figure 3: (a) Statistics of registered vehicle number for Pretoria and South Africa, and (b) National electricty production from 1965 to 1990.

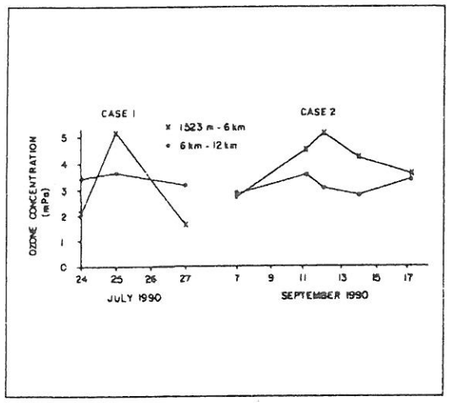

Figure 4: Variation of tropospheric ozone concentration during two ozone episodes'.

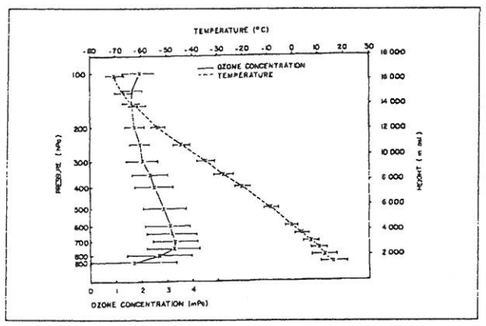

Figure 5: Mean vertical profiles of ozone concentration and temperature in the tropopause above Pretoria. The bars are one standard deviation.

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