THE HAZARDOUS GASEOUS AND VOLATILE CONSTITUENTS WHICH CURRENTLY ACCOMPANY THE PARTICULATES IN THE MAJOR SOUTH AFRICAN URBAN/INDUSTRIAL CENTRES

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SUMMARY

This paper reviews the status as regards the existing levels and trends of the primary inorganic and organic gaseous pollutants occurring in the ambient air of the major South African urban and industrial centres, with specific reference to ambient air quality standards and possible impacts on the local environment.

OPSOMMING

Hierdie referaat gee 'n oorsig van die posisie betreffende die bestaande vlakke en neigings van die primêre anorganiese en organiese gasagtige besoedelstowwe wat in die lug van die vernaamste Suid-Afrikaanse stedelike en nywerheidsentra voorkom, met spesifieke verwysing na lug-kwaliteitstandaarde en moontlike impakte op die plaaslike omgewing.

INORGANIC POLLUTANTS

1. *Fossil fuel related pollutants*

South African cities and towns

Sulphur dioxide arises mainly from the combustion of coal and can be regarded as one of the most common pollutants in South Africa. It is well-known that high concentrations of sulphur dioxide can impair human health.

A large national network covering the major South African urban and industrial centres has been established to determine the levels and the long-term trends of sulphur dioxide pollution levels and from this to assess the efficiency of existing control measures. The measurements are coordinated by the Atmospheric Sciences Division of the CSIR in collaboration with local authorities.

Sulphur dioxide is determined by the acidimetric hydrogen peroxide method[1] which is also employed in the United Kingdom and Europe. This method is relatively simple, fairly sensitive, but suffers from the disadvantages that it is non-specific (it detects other acidic gases as well) and subject to interference from ammonia. However, in general, these gases do not occur in significant amounts in city areas.

The results from the most recent survey (See Table 1) indicated that the annual mean sulphur dioxide concentrations recorded in central city, industrial, and white as well as black residential areas of the major South African cities and towns are well below the ambient air quality standard for annual periods recommended by the World Health Organisation (WHO)[2]. Furthermore, the long-term trends of sulphur dioxide pollution levels calculated for the winter seasons (See Table 2) indicated statistically significant decreases for 36% of the monitoring sites whereas 64% of the monitoring sites showed no significant trends. The majority of those sites displaying decreasing long-term trends (See Figure 1) had been in operation for at least ten years. These decreasing trends can be ascribed to a number of different factors, i.e. a change in the fuel usage pattern by switching from heavy fuel oil to coal in smaller boilers, the wider use of electricity instead of boilers for space heating purposes, the elimination of fire-Places in white residential areas, and control of the release heights of pollution. However, evidence is now emerging that these decreasing trends are beginning to level out as is manifested by the fluctuation of sulphur dioxide concentrations at relatively constant levels and it is quite possible that the greatest improvements in air quality may have been achieved. Examples of such cases are Pretoria – city centre, Cape Town – city centre, Cape Town – Paarden Eiland, Durban – Southern Sewage Works, Pretoria West and Amanzimtoti – industrial (See Figure 1). With regard to the monitoring sites displaying no significant trends, it should be mentioned that these have been operated only for a few years and displayed relatively low sulphur dioxide concentrations.

Eastern Transvaal Highveld

A survey has recently been initiated in the Eastern Transvaal Highveld over an area of about 10 000 km^2 to determine the existing pollution burden in terms of primary and secondary pollutants (including sulphur dioxide) and from this to make projections of future pollution levels with further industrial devleopments in this area. Sulphur dioxide was monitored at different sites (See Figure 2) on a continuous basis by employing Scientific Industries model 67 conductometric analysers. These measurements were done in parallel with wind direction and speed observations.

Preliminary findings obtained from the first survey carried out during the winter season of 1980 indicated that:

- the sulphur dioxide pollution measured in the area originated basically from sources *within* the area. This was manifested by the sulphur dioxide concentration roses recorded at the different monitoring sites (See Figure 2).

- the sulphur dioxide pollution measured in the area originated from both low level sources (nearby towns and/or veld fires) and high level sources (power station stacks). This was manifested by the diurnal concentration patterns displaying early morning as well as midday-early afternoon peak concentrations (See Figure 3); the latter being caused by the downward transportation of sulphur dioxide (originally discharged above the inversion layer) through vertical turbulent mixing resulting from the onset of unstable atmospheric conditions during day time.

- the highest sulphur dioxide concentrations were measured at the Grootpan and Elandsfontein sites (See Table 3). However, none of the hourly or daily concentrations measured at these or any of the other sites (See Table 3) exceeded the most stringent relevant ambient air quality standards[2,3].

These results should be regarded as preliminary and future monitoring of sulphur dioxide levels should be carried out over a larger area in the Eastern Transvaal Highveld in order to obtain a more realistic assessment of the pollution burden situation. In addition, it will be essential that a careful study be made of the atmospheric chemistry of sulphur dioxide particularly as regards its transformation in and removal from the atmosphere under the climatic conditions of the Eastern Transvaal Highveld.

2. *Traffic related pollutants*

Motor vehicles are the primary sources of carbon monoxide, hydrocarbons and oxides of nitrogen in urban centres. Moreover, hydrocarbons and nitrogen oxides are known to be the precursors of oxidants and have caused considerable public discomfort in cities like Los Angeles, New York and Tokyo.

The traffic-dense areas of Pretoria, Johannesburg and Durban have been surveyed regularly since 1968 to establish the trends of traffic pollution levels and from this to evaluate the necessity of introducing control measures in South Africa. Two air-conditioned mobile laboratores equipped with continuous analysers were used to carry out these surveys. Instrumentation in these mobile laboratories comprised:

- Beckman model 865 nondispersive infra-red carbon monoxide analysers fitted each with molecular sieve scrubbers to eliminate interference from atmospheric water vapour

- Beckman model 400 flame ionisation total hydrocarbon analysers

- Beckman model 952 chemiluminescent ozone analysers

- strip chart recorders to obtain a permanent record of all the measurements from the continuous analysers.

Regular on-site calibration of all the monitoring equipment was carried out using certified gas standards and in the case of ozone analysers, an ozone generator, the output of which was checked with the standard neutral potassium iodide spectrophotometric method.

The diurnal concentration patterns recorded for carbon monoxide, hydrocarbons and nitrogen oxides were found to be very similar, in that maximum concentrations were displayed which coincided with peak traffic periods (See Figure 4). Likewise, the weekly concentration patterns for these pollutants were found similar in that generally a build-up of pollution levels occurred during the week to reach a maximum towards Friday, followed by a minimum over weekends (See Figure 4). This was possibly related to the prevailing traffic-flow patterns in all three of the cities. However, the situation was quite different for ozone which is known to be produced photochemically from the reaction between nitrogen oxides and unburnt petrol vapours. The diurnal concentration patterns for ozone usually displayed a gradual increase in concentration during the day to reach a maximum plateau value just past midday (See Figure 4), this being directly the result of the photochemical reaction between nitrogen oxides and unburnt petrol vapours. The weekly concentration patterns indicated that slightly elevated ozone levels were reached during weekends (See Figure 4). This phenomenon was probably due to the scavenging of ozone by nitric oxide[4] which is more pronounced during the week when nitric oxide emissions are higher. In general, it appeared that the peak concentrations recorded for traffic pollutants during diurnal periods were too low and of too short a duration to cause any serious health effects to the average individual.

The results from the most recent survey (See Table 4) indicated that the mean daily concentration of traffic pollutants recorded during the winter season for Pretoria, Johannesburg and Durban compared favourably with the ambient air quality standards for 24-hour periods accepted for city atmospheres[2,5 - 10]. A comparison of individual daily concentrations with the ambient air quality standards for 24-hour periods (See Table 5) showed that for carbon monoxide and ozone the relevant ambient air quality standards were practically never exceeded whereas for nitrogen oxides and nitrogen dioxide the relevant ambient air quality standards were exceeded on a fairly large number of days in Pretoria. In addition, special investigations in Pretoria and Durban[11, 12] showed that higher concentrations of ozone occurred in suburban areas than in city-centres; the mean daily ozone concentrations in suburban areas being about double those in city centres. This is probably due to the scavenging of ozone by nitric oxide[4] which is present in much higher concentrations in city-centres. This finding indicated the importance of monitoring ozone pollution levels also in suburban areas of cities.

The trends of the mean daily traffic pollution levels for the winter season (See Figure 5) indicated decreasing tendencies for carbon monoxide and hydrocarbons but increasing

tendencies for nitrogen oxides in the three cities of Pretoria, Johannesburg and Durban. In the case of ozone increasing tendencies were indicated for Pretoria and Johannesburg but no significant trend was displayed for Durban. These trends can be ascribed to a number of different factors, i.e. the introduction of one-way traffic systems in cities, improved carburettor as well as engine design features and, more recently, the running of engines on leaner air/fuel mixtures to conserve fuel. All of these factors promoted a better combustion of fuel hence resulting in lower emissions of carbon monoxide and hydrocarbons but in a higher emission of nitrogen oxides. The higher levels of nitrogen oxides, in turn, lead to an increased production of ozone via the pathway of photochemical reactions. Furthermore, it is of relevance to point out here that, provided the traffic flow patterns and the total traffic flows do not change too drastically in the future in the cities of Pretoria, Johannesburg and Durban, it is not unlikely that the existing trends for carbon monoxide, hydrocarbons and nitrogen oxides might eventually level out. However, the existing increasing trend for ozone might continue because the introduction of alcohol-blended petrol in South Africa has enhanced the potential for the formation of photochemical smog and hence ozone due to the resulting concurrently higher emission of oxygenated hydrocarbons.

The findings reported here for Pretoria, Johannesburg and Durban as well as those reported in an independent survey for Cape Town[13] indicate that

- It is imperative that a regular monitoring of traffic pollutants, in particular nitrogen oxides and ozone, should be carried out in the city-centres as well as suburban areas of major South African cities. Also techniques should be developed to monitor pollutants such as oxygenated hydrocarbons, the emission of which will be enhanced by the addition of alcohol to petrol in South Africa

- the potential for photochemical smog occurrence in the cities of Pretoria, Johannesburg and Durban has increased as is manifested by the increasing trends of the levels of nitrogen oxides and ozone.

3. *Industry-related pollutants*

Fluorides (both gaseous and particulate) are fairly commonly occurring industry-related pollutants. They are released by various industrial processes such as phosphate fertilizer production, aluminium reduction, metal smelting processes and ceramics manufacturing.

Because of the known adverse effects associated with fluorides considerable attention has been given to their measurement and occurrence in a number of different urban and industrial areas of South Africa. Sampling and analysis of gaseous fluorides[14] were accomplished by collection on sodium formate impregnated filters preceded upstream by untreated filters to remove particulate fluorides, a clear-cut separation of the collected gaseous and particulate fluorides by microdiffusion, aqueous extraction of the samples and, finally, determination of the fluorides by the fluoride ion-selective electrode.

Examples of typical surveys of fluorides that have thusfar been undertaken included:

- the determination of the pollution burden in terms of fluorides (and other gasesous pollutants such as sulphur dioxide) in the neighbourhood of fertilizer complexes to establish the exposure hazard to nearby town inhabitants and from this the need to implement stricter control measures

- determination of the effects of fluorides on sugar cane plants at levels that were expected to occur in the neighbourhood of a to-be-erected aluminium smelter to assess the efficiency of the proposed control measures

- the determination of background levels of fluorides in the atmosphere and vegetation in the vicinity of an oil refinery complex prior to the commissioning of a hydrogen fluoride alkylation plant. Once this plant becomes operative the impact on the immediate environment can be assessed.

These surveys served to provide important information on the actual or potential impact of fluorides within these environments. However, because of the confidential nature of this work, no detail of the actual results can be presented in this paper.

ORGANIC POLLUTANTS

Volatile organic substances are released by various industrial processes as well as by motor vehicles. Some of these organic substances are known to be either potent eye irritants or basically toxic in nature, whilst others are known to be the precursors of photochemical smog.

In order to permit efficient control of the emission of organic substances it is essential that their chemical nature should be known as well as that of those products which may be formed as a result of chemical reactions in the atmosphere between these compounds and other air pollutants. Consequently, two monitoring systems have been developed i.e. one for the scanning of city and industrial atmospheres for a relatively broad spectrum of volatile organic substances and another for the routine determination of a number of specific major volatile organic substances in city air. The procedures[14 - 23] involved collection of air samples on adsorbents (chromosorb and activated charcoal), sample recovery by either heat desorption (chromosorb) or solvent extraction (activated charcoal) and analysis by capillary gas chromotography/flame ionisation detection/mass spectrometry as well as packed column gas chromatography/flame ionisation detection.

Surveys conducted during 1975/76 and 1979 showed that no 'unexpected' volatile organic substances occurred in the air of Pretoria, Johannesburg and Durban. The organic substances identified were constituted mainly of paraffinic, olefinic and aromatic hydrocarbons with only a few being oxygenated and chlorinated hydrocarbons. Moreover, it was found that no basic chemical changes have apparently taken place in terms of the C_6 to C_{13} volatile organic substances in the atmospheres of these areas over the $\frac{3}{4}$ year period that elapsed between the two surveys; the same C_6 to C_{13} organic substances being identified during both surveys.

In addition, diurnal and weekly concentration patterns of a number of major volatile organic substances were established during a brief survey carried out in the winter of 1980 in the city-centre of Johannesburg. These patterns (See Figure 6) displayed the same features as those for carbon monoxide, total hydrocarbons and nitrogen oxides (See Section 2) in that peak concentrations coincided with peak traffic periods.

Finally, the most recent quantitative measurements carried out during the winter season of 1979 indicated that the daily concentrations of the major C_6 to C_{13} volatile organic substances (See Table 7) were well below the accepted ambient air quality standards[24] for city and industrial areas. Furthermore, judging by the relatively low concentrations of photochemically produced substances, e.g. ozone, recently measured in the air of Pretoria, Johannesburg and Durban, (See Section 2), it appears that the volatile organic substances occur in concentrations which are apparently still too low to produce any significant photochemical reactions in the air of these three cities. However, this does not seem to be the case for Cape Town where recently relatively high ozone levels have been recorded[13].

CONCLUSIONS

The sulphur dioxide pollution levels occurring in the major South African urban and industrial centres are well below the ambient air quality standard accepted for city atmospheres. The long-term trends of sulphur dioxide pollution levels still show decreases in a number of city-centre and industrial areas but there is evidence that these trends are beginning to level out and it is possible that the best possible improvement in air quality has been reached. Moreover, the possibility that sulphur dioxide levels might be increasing in the near future due to a continued expansion of industrial activity cannot be excluded.

The daily levels of carbon monoxide and ozone recorded in Pretoria, Johannesburg and Durban practically never exceeded the relevant ambient air quality standards. However, the daily levels of nitrogen oxides recorded in Pretoria exceeded the relevant ambient air quality standards in a fairly large number of cases. In addition, there is evidence for increasing trends in the levels of nitrogen oxides and ozone, hence indicating that the potential for photochemical smog formation in these cities has increased. In Cape Town ozone levels exceeding the relevant ambient air quality standards have, in fact, already been recorded. Furthermore, arising from discussion in the literature, it appears that the introduction of alcohol-blended petrol in South Africa will enhance the potential for the formation of photochemical smog in city-centres. Consequently, the situation as regards traffic related pollutants does present some reason for concern and it is imperative that these pollutants be regularly monitored in order to assist the control authorities in making a timely and well-motovated decision as regards the control of motore vehicle exhaust emissions in South Africa.

A wide spectrum of potentially harmful volatile organic substances occur in the air of South Africa urban and industrial centres. However, these organic substances are present in still too low concentrations to either cause any serious undesirable health effects to the average individual or to significantly promote photochemical reactions in South African city atmospheres.

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Table 1

Sulphur Dioxide Concentrations in the Air of SA Cities and Towns

City/Town	Annual mean sulphur dioxide concentrations ($\mu g/m^3$)			
	Central	Industrial	Residential (white)	Residential (black)
Cities				
Bloemfontein	33	37	7	30
Cape Town	20	23	9	–
Durban	26	39	11	–
Kroonstad	–	20	–	–
Port Elizabeth	18	20	15	8
Pretoria	17	21	10	–
Springs	32	33	–	28
Towns				
Amanzimtoti	4	11	–	–
Edenvale	15	11	–	–
Estcourt	10	–	–	–
Newcastle	18	15	15	–
Witbank	8	7	8	–

NOTE:

(i) Annual means were calculated over the last three years of survey, i.e. 1978, 1979 and 1980 to obtain a statistically valid annual mean.

(ii) Ambient Air Quality Standard for annual periods[2]: 60 $\mu g/m^3$ (WHO, 1972)

Table 2

Long-term Trends of Sulphur dioxide Pollution occuring in SA Cities and Towns

City/Town	Number of sites	Trends		
		D	I	O
Cities				
Bloemfontein	4	1	–	3
Cape Town	7	3	–	4
Durban	9	2	–	7
Kroonstad	1	–	–	1
Port Elizabeth	6	–	–	6
Pretoria	4	4	–	–
Springs	3	2	–	1
Towns				
Amanzimtoti	1	1	–	–
Edenvale	2	1	–	1
Newcastle	2	1	–	1
Witbank	3	–	–	3
Totals	42	15	–	27
Percentages		36		64

NOTE:

(i) D : decrese
I : increase
O : no significant trend

(ii) Long-term trends were calculated for monitoring sites which had been in operation for four or more *winter seasons.* Long-term trends were obtained by the technique of linear regression. A trend indicated a decrease or increase if the probability that the relevant tendency continued was higher than 90% If this was not the case, the trend was considered to be not significant.

Table 3

Traffic Pollutant Concentrations in the Air of SA Cities

City	Season	Mean daily concentrations				
		CO ppm	HCS ppm	NO_x pphm	NO_2 pphm	O_3 pphm
Pretoria	Winter 1980	5,4	1,9	31,7	11,2	0,6
Johannesburg	Winter 1980	5,0	1,4	11,2	4,2	1,1
Durban	Winter 1979	2,1	–	17,8	6,5	0,3
	Winter 1980	–	–	–	–	0,7

NOTE:

(i) Measurements were carried out at one traffic-dense site per city.

(ii) CO: carbon monoxide; HCS: hydrocarbons, expressed as methane; NO_x: nitrogen oxides, expressed as nitrogen dioxide; NO_2: nitrogen dioxide, calculated from measured nitrogen oxides and nitric oxide concentrations; O_3: ozone.

(iii) Ambient Air Quality Standards for 24-h periods are:

CO:	8 ppm	(VDI Commission, West German, 1967)[5]
	10 ppm	(Environmental Council of Japan, 1970)[6]
NO_x:	32 pphm	(Israel)[7]
NO_2:	11 pphm	(Department of Environment, Canada, 1971)[8]
	13 pphm	(Environmental Protection Agency, USA, 1971)[9, 10]
Oxidants (as O_3):	2,5 pphm	(Department of Environment, Canada, 1971)[8] (VDI Commission, West Germany, 1974)[2]

Table 4

Daily Traffic Pollutant Concentrations vs AAQS

City	Season	Days exceeding AAQS (%)					
		CO		NO_x	NO_2		O_3
		8 ppm	10 ppm	32 pphm	11 pphm	13 pphm	2,5 pphm
Pretoria	Winter 1980	6	0	47	53	26	0
Johannesburg	Winter 1980	0	0	0	0	0	0
Durban	Winter 1979	0	0	11	0	0	0
Wint	Winter 1980	–	–	–	–	–	0

NOTE:
(i) AAQS: Ambient Air quality Standards for 24-h periods (see Footnote, Table 3)
(ii) CO: carbon monoxide; No_x: nitrogen oxides, expressed as nitrogen dioxide; NO_2: nitrogen dioxide

Table 5

Volatile Organic Substance Concentrations in the Air of SA Cities

Organic substances	Daily concentrations (ppb)													
	Pretoria		Johannesburg		Durban		Pretoria		Johannesburg		Durban			
	C		C		C		I		I		I (1)		I (2)	
	A	B	A	B	A	B	A	B	A	B	A	B	A	B
Paraffinic hydrocarbons														
n-Octane	2,4	3,7	0,5	1,3	0,1	0,1	0,7	1,3	0,7	1,3	0,3	0,7	0,5	1,1
n-Nonane	1,5	2,1	0,4	0,9	0,1	0,1	0,4	0,7	0,6	1,3	0,8	1,8	0,8	1,8
n-Decane	1,3	2,7	0,3	0,6	0,1	0,1	0,3	0,5	0,6	1,2	0,5	1,2	0,7	1,4
Aromatic hydrocarbons														
Benzene	6,5	12,2	1,8	4,7	0,5	0,9	2,7	4,6	2,5	4,8	0,8	1,6	0,8	1,3
Toluene	10,9	22,6	2,9	8,3	1,2	1,7	7,3	17,5	13,7	24,1	4,2	9,7	8,2	17,7
Ethylbenzene	2,5	4,0	0,7	1,8	0,3	0,4	1,1	1,9	1,1	1,9	0,7	1,4	1,6	3,3
o-, m-, p-Xylenes	7,9	16,6	2,4	5,7	1,3	1,5	3,1	4,3	3,7	6,2	2,5	5,0	5,1	8,9
n-Propylbenzene	1,1	2,5	0,2	0,4	0,1	0,1	0,2	0,3	0,2	0,3	0,2	0,4	0,2	0,3
1-Methyl-3-ethyl-benzene + 1-Methyl-4-ethylbenzene	4,8	12,6	0,7	1,5	0,3	0,4	0,3	0,6	0,6	1,1	0,5	0,9	0,7	1,4
1, 3, 5-Trimethyl-benzene	1,6	3,8	0,2	0,5	0,1	0,2	0,2	0,3	0,3	0,7	0,1	0,3	0,3	0,6
1, 2, 4-Trimethyl-benzene	5,7	13,6	0,7	1,5	0,3	0,5	0,4	0,8	0,6	0,9	0,9	1,4	0,8	1,7
Chlorinated hydrocarbons														
Tetrachloro-ethylene	2,3	6,0	–	–	–	–	1,1	2,5	0,4	1,0	–	–	–	–

NOTE:
(i) Measurements were made during the winter season of 1979 (18.06.79 – 3.08.79)
(ii) ppb: parts per 10^9; C: city centre; I: industrial area; A: mean daily concentration; B: maximum daily concentration
(iii) Accepted concentration limits[24] for volatile organic substances are:

n-Octane	: 6 000 ppb (C),	300 ppm (I);
n-Nonane	: 4 000 ppb (C),	200 ppm (I);
Benzene	: 200 ppb (C),	10 ppm (I);
Toluene	: 2 000 ppb (C),	100 ppm (I);
Ethylbenzene	: 2 000 ppb (C),	100 ppm (I);
Xylene	: 2 000 ppb (C),	100 ppm (I);
Trimethylbenzene	: 500 ppb (C),	25 ppm (I);
Tetrachloroethylene	: 1 000 ppb (C),	30 ppm (I).

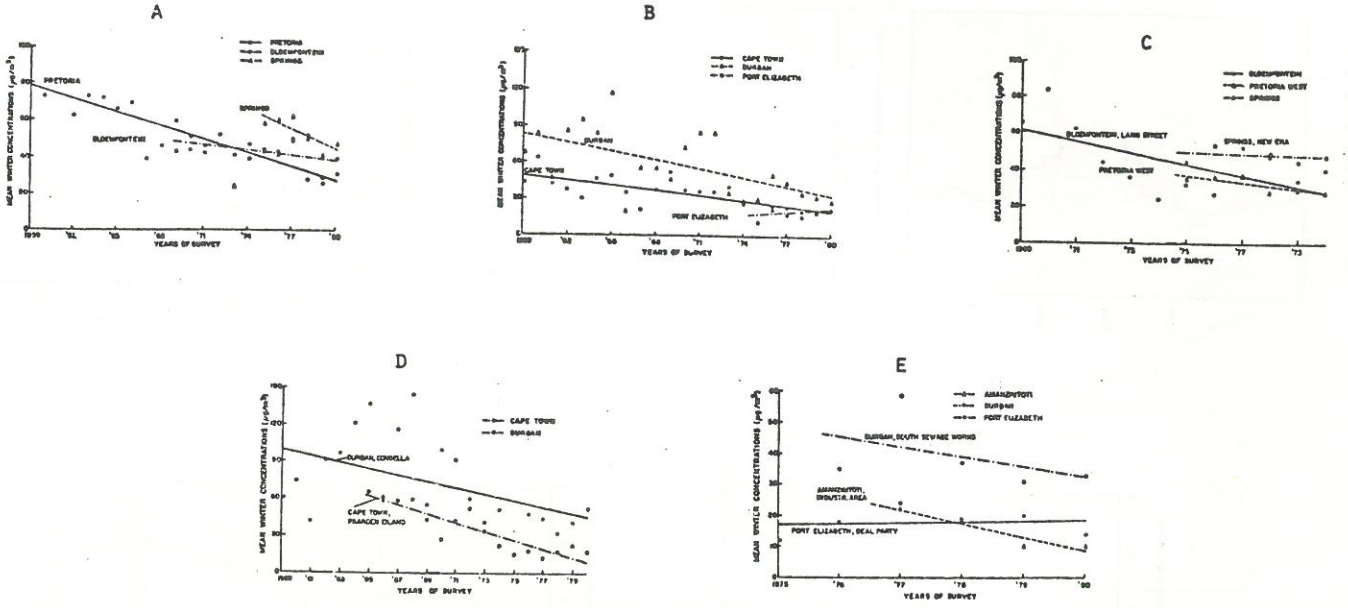

FIGURE 1 Long-term trends of sulphur dioxide pollution levels recorded in SA city-centres (A, B) and industrial areas (C, D, E) for winter seasons.

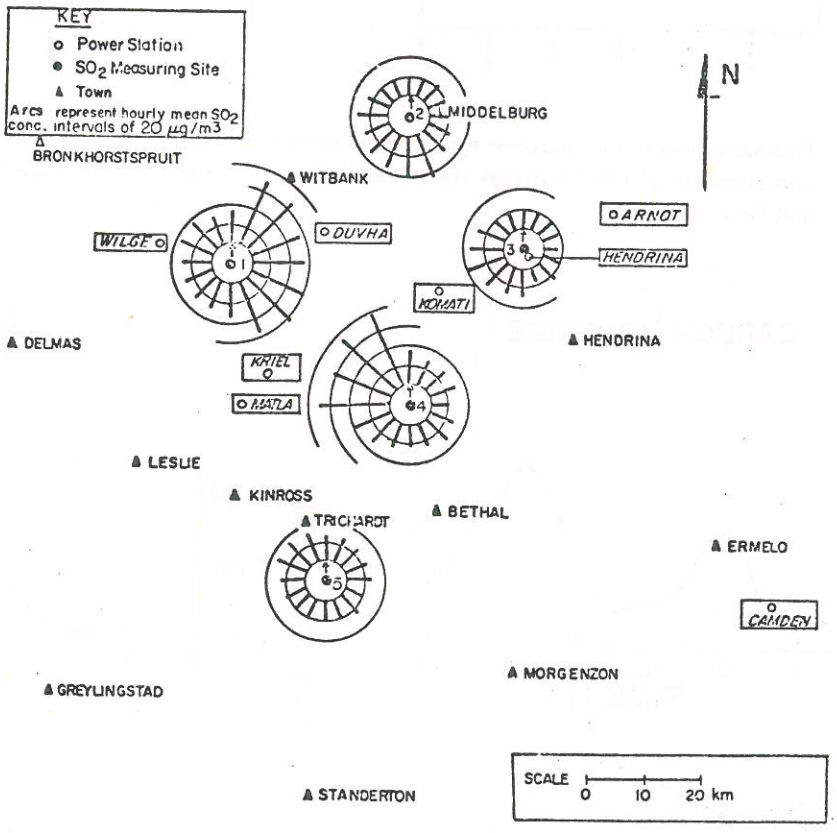

FIGURE 2 Sulphur dioxide concentration roses recorded in the Eastern Transvaal Highveld for the winter season of 1980. Measuring sites are at Grootpan (1), Middelburg (2), Hendrina (3), Elandsfontein (4) and Secunda (5).

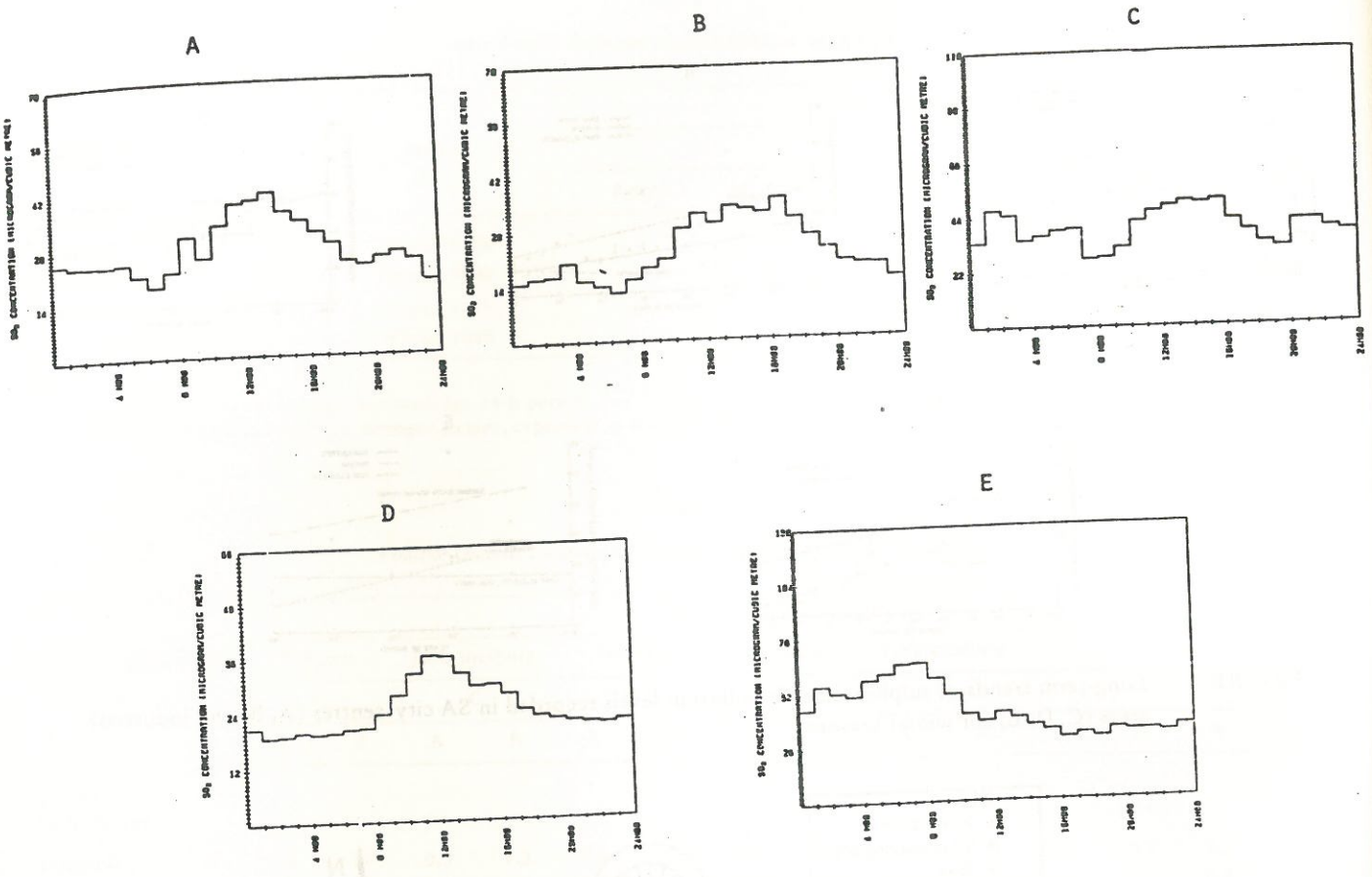

FIGURE 3 Diurnal concentration patterns for sulphur dioxide recorded in the Eastern Transvaal Highveld for the winter season of 1980. Patterns are for Middelburg (A), Hendrina (B), Elandsfontein (C), Secunda (D) and Grootpan (E).

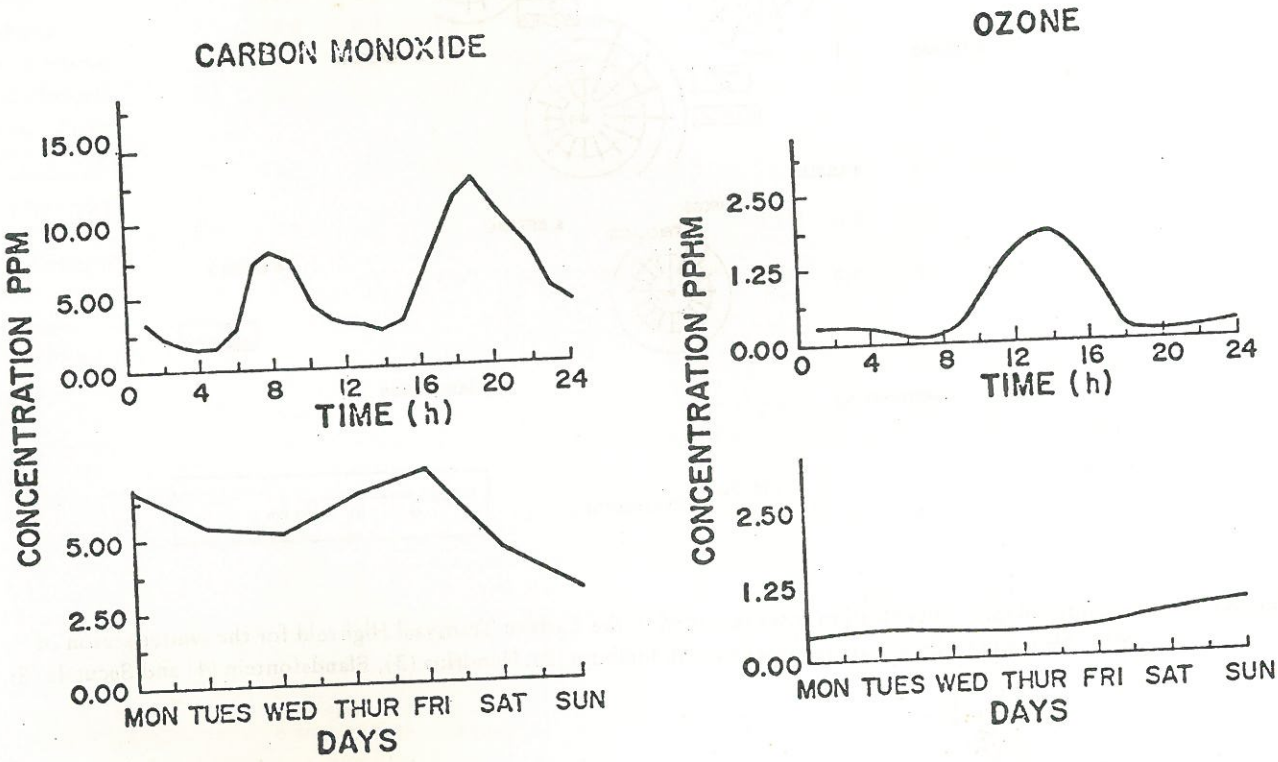

FIGURE 4 Typical examples of diurnal and weekly concentration patterns for carbon monoxide and ozone recorded in SA city centres.

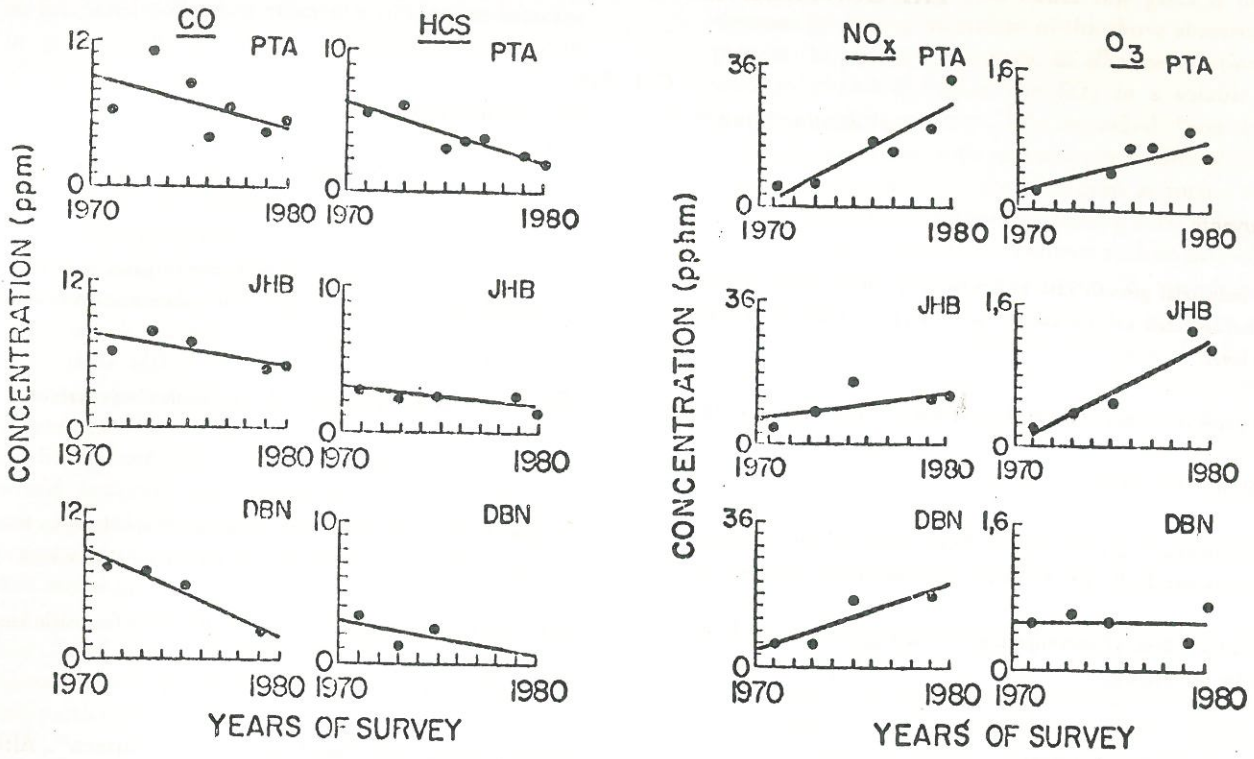

FIGURE 5 Trends of traffic pollution levels recorded in SA city-centres for winter season. Pollution data presented are for carbon monoxide (C0), hydrocarbons (HC), nitrogen oxides ($N0_x$) and ozone (0_3). Curves were fitted to the data points by linear regression.

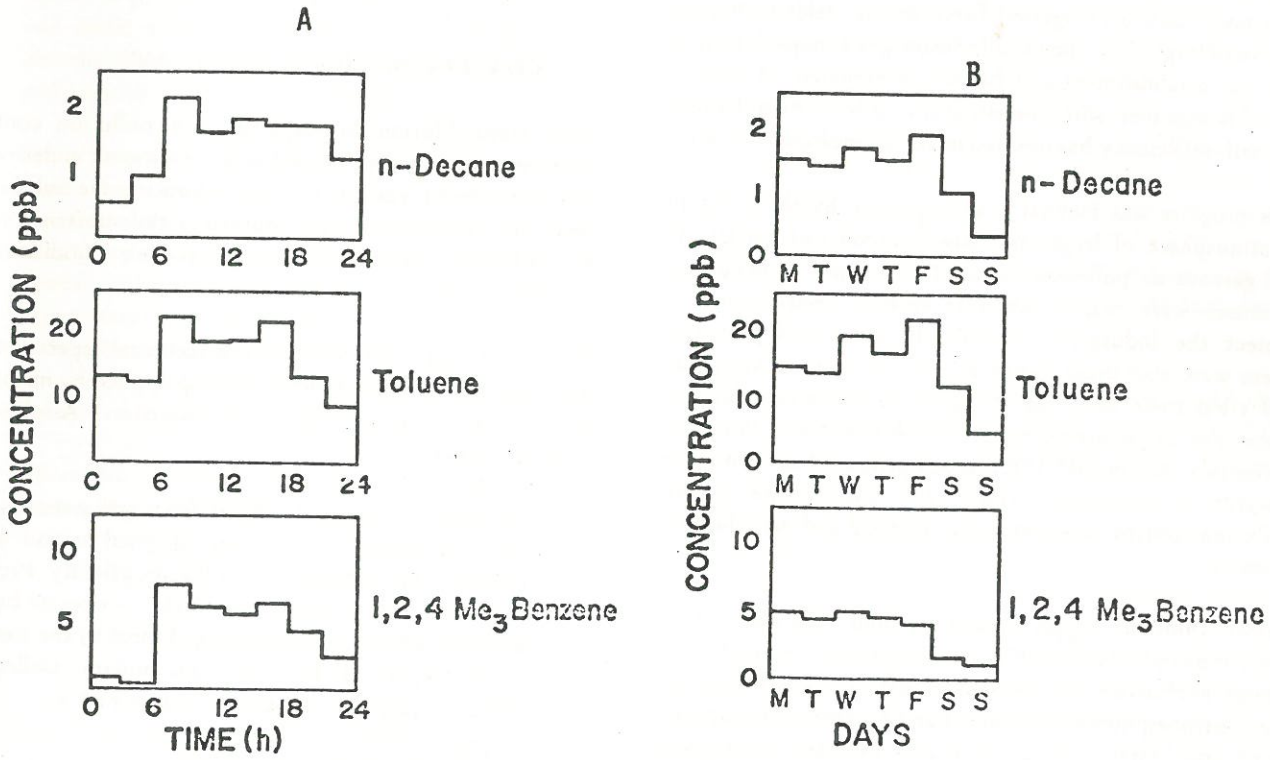

FIGURE 6 Diurnal concentration patterns (A) and weekly concentration patterns (B) for a number of major volatile organic substances recorded in the city-centre of Johannesburg for the winter season of 1980.