

PARTICULATES IN URBAN AND INDUSTRIAL ATMOSPHERES OF SOUTH AFRICA

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SYNOPSIS

Since the inception of an Air Pollution facility at the CSIR a survey of smoke has been in operation, first in Pretoria, and, with the passage of time, in a large number of towns. A survey of trace elements was started up in 1976 and has also been extended to a number of towns. This paper describes the nature of the measurement techniques and the results which have been obtained.

OPSOMMING

Sedert die ontstaan van 'n Lugbesoedelingsnavorsingsfasiliteit by die WNNR is rookopnames gedoen, eersens in Pretoria en daarna in 'n groot aantal dorpe. Daar is in 1976 begin met 'n opname van spoorelemente wat ook na 'n aantal dorpe uitgebrei is. Hierdie referaat beskryf die aard van die meettegnieke en die resultate wat verkry is.

1. SMOKE AND CARBONACEOUS PARTICLES

1.1 *Measuring Technique.* The measurement of the quantity or the concentration of smoke in a way suitable for continuous surveys presents some problems because in general, the particulates suspended in the atmosphere consist of particles of different shapes, sizes and chemical compositions and because the heterogenous mixture differs greatly in its characteristics from one location to another and from one occasion to another at any one site. While techniques have advanced to the extent that certain size fractions can now be collected by means of cyclones, impactors and elutriators, it is not possible to collect the particles selectively according to their chemical composition.

The 'filter paper method' which is standardized in South Africa¹ was evolved by the National Physical Research Laboratory in the CSIR with the aim to be specific for the dark particles which are emitted by combustion processes and remain suspended in the atmosphere over long periods. The goal was achieved by

1. operating with sampling velocities smaller than 3 cm/s ($3 \cdot 10^{-2} \text{ms}^{-1}$) which ensures that the deposition of particle sizes smaller than 10 μm , and by
2. determining the quantity of the particulate sample of the filter by measuring the light absorbed by them in recognition of the optical characteristic that the extinction by coal and carbon particles is as much as ten times greater than the extinction by transparent mineral particles².

The filter paper method is described in previous publications^{3, 4, 5}.

1.2 *Air Quality Standards for Smoke.* The difficulties encountered when measuring smoke are reflected in the way the setting of air quality standards is handled in various countries. The USA⁶ laid down air quality standards for 'total suspended particulates' in size ranges between 0,1 and 100 μm , but none for smoke, which may be due to the fact that the American measuring method for smoke by means of automatic recorders has some shortcomings. It assumes a linear relationship between the quantity of the dark particles on the filter and the light transmitted by them when, in fact an equation of the form $Q = a \times D_3^5$ where D is the light extinction, would describe the relationship better.

The World Health Organization⁷ in 1972 set standards for suspended particulates' smaller than about 10 μm , which are the particles of concern to public health, giving as reference method the British filter paper method which was developed for the measurement of 'smoke'. In 1979 after further cooperative studies with European research organisations the WHO⁸, still referring to particles smaller than 10 μm , stressed that 'smoke' and 'suspended particulates' are two different entities and defined

1. 'smoke, assessed in terms of blackness, as a measure of pollution associated with the incomplete combustion of fuel' and
2. 'total suspended particulates, determined by weight, as a wider concept that includes all material which, by virtue of its particle size, remains in suspension for long periods'.

In the same year the WHO 1. set new guidelines for 'smoke' and 2. recommended tentative guidelines for 'suspended particulates measured by weight' which are given in Table II. The standards for smoke are 50% higher than those set in 1972.

1.3 Smoke Levels in South African Urban Atmospheres.

Smoke is monitored at 130 sampling sites situated in 32 cities and towns. Usually three filter units are in operation, one being installed in the central, one in industrial and one in a residential area. The lack of the selectivity of the sampling process is considered by *not* installing the sampling instruments in the vicinity of specific industries, but also not close to a prominent smoke source. The public health aspects are taken into account in that the instruments are mounted about two metres above the ground, and frequently are installed at places where people move or gather.

The results which the extensive survey in South Africa supplied are as follows:

- a) At every sampling site *smoke pollution shows two distinctive types of behaviour* which are characterized by a six-month period (October to March) during which the concentrations are low and a six-month period during which the concentrations 1. increase (April, May), 2. reach their maxima (June, July) and 3. decrease to the original low levels (August, September)^{9, 10}. The second six-month period coincides with the winter season in South Africa and represents the period, which, from the point of air pollution control demands careful attention.

The variation in the smoke levels is caused more by climatic factors like rainfall, wind speed and, most important of all, atmospheric stability than by human activities. Atmospheric stability is pronounced on the Highveld plateau where 68% of the monitoring sites are situated, because nocturnal surface inversions, varying in depth between 300 and 400 m, occur during the winter, with strengths of 2 to 6°C on the average and with the maxima as high as 20°C¹¹. The frequency of the occurrence of the radiation inversions is around 80% during the winter and 40% during the summer season.

- b) The *smoke levels are different* at various monitoring sites as can be seen from the annual mean concentrations, set out in $\mu\text{g}/\text{m}^3$ in Table III. The figures in the column A represent the means of the last three years of the survey (1978 to 1980) while the figures in column B represent the annual mean concentrations of the first three years of the surveys which may have started in any year between 1958 and 1974. The levels depend on source factors such as emission strength and height and on the meteorological factors prevailing over the different geographical areas. The wind speeds are remarkably lower on the Highveld than in the coastal region¹² which consequently influences the efficiency of the pollution transport and the mixing and removal mechanism over the various urban regions.

The monitoring sites in the interior of South Africa therefore show higher smoke concentrations than the corresponding sites in the coastal regions. As a matter of fact, Pretoria where South Africa's air pollution research started, has atmospheric conditions very unfavourable for the dispersal of pollution which leads to smoke concentrations being higher than in other comparable cities¹³.

- c) As far as the *long-term trends* are concerned, Table III shows that in the central and industrial areas of the cities and towns which started monitoring before 1972, the smoke levels have decreased to between 40 and 60% of their initial levels; where monitoring started after 1972 the percentage decrease is between 10 and 35%. In the residential areas the decrease in the concentrations is dependent on the original levels: where these were high, as in the case of Pretoria, the decrease is 56% while in the coastal cities of Cape Town and Durban where the levels were always low, no decrease has occurred. The figures for the black residential areas, in Bloemfontein and Port Elizabeth indicate that the smoke levels have decreased by 25%.

The long-term trends of the main industrial areas and of two mixed industrial-residential areas are graphically presented in Figures 1 to 3 proving that at most of the sampling sites the smoke concentrations have decreased quite significantly.

- d) The improvement in the air quality is, with regard to the centre of Pretoria, confirmed by the estimates of horizontal visibility made since 1966 by the South African Weather Bureau, situated in this part of the city (Figure 4). The observations made at 08h00 during June 1966 indicate that the lowest visibility estimates referred to distances smaller than 100 m (frequency 3%) and the best referred to a distance of 8 km (frequency 3%) whereas in June 1979, at 08h00, the respective distances were 800 m and 85 km and the respective frequencies were 10 and 3%. Looking at the curves in Figure 4 it appears that in the centre of Pretoria the visibility improved mainly between 1970 and 1975 when the lowest visibility estimates increased from 100 m and smaller to 400 m, while a general improvement over the whole city occurred between 1975 and 1979. This observation is in excellent agreement with the monitoring results which show that between 1966 and 1971 the smoke level in the centre of Pretoria hardly changed while between 1971 and 1974 they decreased at about 15% per year. They decreased further until 1979 to reach a level which is about 40% of the 1971 level.

It is certain that in other cities and towns the reduction in the smoke levels is also accompanied by an improvement in the visibility but not every town has a weather station in the vicinity of the sampling site to prove it.

1.4 *Assessment of the Smoke Levels.* A review of the South African smoke levels in terms of the new guidelines of the WHO which set as the standard an annual average concentration (arithmetic mean) of between 40 and 60 $\mu\text{g}/\text{m}^3$, gives a more favourable picture than that obtained two years ago when the pollution situation was last reviewed¹⁴. The reason for this is not the raising of the standard by the WHO but the continuation of the decreasing trend at many of the sampling sites. At the present time the smoke standard is exceeded in cities where the ventilation potential is poor (Pretoria, Bloemfontein) or in the cities and towns where a smoke source bigger than the urban service industries is located (Cape Town, Boksburg) or where railway shunting yards are situated (Springs). The standard is also exceeded in some of the black townships on the Highveld (Bloemfontein, Kroonstad).

The statistical examination of the long-term trends at the sampling sites which exceed the standards indicates that a reduction of the smoke levels can be expected with a 90% probability in the centre of Pretoria, the industrial site in Cape Town and at the industrial and black residential sites of Bloemfontein. No forecasts can be made for other stations.

1.5 *Assessment of the Levels of Total Suspended Particulates.* The new guidelines of the WHO for 'total suspended particulates as determined by weight' make it possible to assess the particulate samples collected at sampling sites where the conversion factor could not be determined because the natural dust load was high. An example of the levels of TSP found in the centre of a town on the Highveld, where the central area not only houses shops and office blocks but has sportfields, and where specific dust producing industries are situated on the periphery, is given in Figure 5. The limits of the WHO which allow concentrations of between 150 and 230 $\mu\text{g}/\text{m}^3$ for a 24-hour sampling period, may not always be fulfilled, but not, as Figure 5 shows, because the concentrations of smoke and trace elements are high, but because of the fraction which is not analysed and which very likely is made up of silicon and other crustal elements.

2. TRACE ELEMENTS IN PARTICULATES IN URBAN AND INDUSTRIAL ATMOSPHERES OF SOUTH AFRICA

Smoke and sulphur dioxide have been measured in South African cities for many years so that, in spite of seasonal variations, long term trends can be observed. In contrast, trace elements have only been measured since 1975 or 76

at most of the sampling sites jointly run by the CSIR and the AEB. The observed concentrations are also strongly seasonal and it is still too early to state whether trends observed so far have long term significance or are simply the result of the variability of the weather. For example Figure 6 shows the seasonal nature of Pb in the air at a central Cape Town site over a period of four years¹⁵. The measurements are made on monthly samples, shown by the dotted line. The solid line is the three point moving average which is useful in smoothing the results. No clear trend is discernable from either presentation. If it is too soon to be sure of trends what can be learnt at this stage?

MAN-MADE POLLUTION VS NATURAL DUST

The sampling network has background or at least rural sampling sites at Cape Point, Table Mountain, Langebaan and Sutherland. At these sites natural aerosol sources such as sea spray and soil dust can be expected to predominate. Results from these sites form a basis for comparison with the results obtained in urban or industrial sites.

In order to determine the significance of the various concentrations as measured at the different sites and to distinguish between probable sources of airborne particulate material, elemental ratios in the aerosols in ambient air and the ratios in possible sources may be compared. By normalisation the concept of relative element enrichment, now in general use, can be derived, whereby the enrichment in air of a given trace element relative to, for example, soil-derived Sc, is determined from the ratio

$$\frac{\text{trace-element concentration in air}/\text{Sc concentration in air}}{\text{trace-element concentration in soil}/\text{Sc concentration in soil}}$$

This concept would imply that if the enrichment is unity or close to unity, the source of the trace element in the aerosol is the medium used in the normalisation, i.e. soil in the above example. Conversely, if the element enrichment relative to a natural source is substantially larger than unity, a different source, probably anthropogenic, is indicated. The concept of enrichment would ideally require that the elemental composition of the donor soil be known, and that the reference element in air, i.e. Sc, be soil-derived rather than an anthropogenic pollutant. While the latter assumption regarding Sc has fairly general validity, the elemental composition of the donor soil presents a very real problem in that it may vary significantly for different areas. However, it has been demonstrated¹⁶ that average global soil values¹⁷ can be used with great success.

At Sutherland in 1977/78 the Er values were as shown below:

Site	Al	Br	Ca	Cd	C1	Co	Cr	Cs	Cu	Eu	Fe	K	Mg	Mn	An	Ni	Pb	Rb	S	Sb	Sc	So	Ti	V	Zn
Sutherland	0,45	30	0,86	610	340	1,44	0,66	0,66	5,2	0,91	0,50	0,54	1,6	0,48	4,3	7,3	36	0,84	8,7	0,84	1,0	41	0,42	1,21	9,4

The elements expected to be typical components of soil dust i.e. Al, Ca, Cr, Fe, Eu, K, Mg, Mn, Rb and Ti show enrichments close to unity. Some enrichments such as those for Br (30x), Cd (610x), Se (41), Zn (9,4x) depend on measurements of low concentrations and so have a greater analytical uncertainty. It has been suggested¹⁸ that instead of using crustal soil or rock compositions as a reference for the purpose of determining element enrichments, the composition of aerosols sampled in remote areas could be used. This has the advantage of more reliably accentuating anthropogenic enrichments but this method does not contribute to the identification of natural enrichment processes nor low-level global pollution.

The Er shows up the anthropogenic origin of elements such as Pb and Br very clearly but can be deceptive if Sc, the normalising element is liberated significantly by pollution sources. For example at a Pretoria West site close to Iscor an average concentration of $22 \mu\text{g Fe/m}^3$ was observed in 1977/78 but the Er was only 2,2x. Clearly Sc is also a component of the Iscor emissions thus depressing the Er for iron and other elements. In cases such as this an element other than Sc should be used for normalisation. When urban aerosols are weighed prior to analysis it is typically found that the weight of the elements analysed for, totals between 1 and 10% of the aerosol sampled Figure 5. The remainder consists of elements such as carbon, oxygen, and silicon which are not usually determined. This result is also found when deposited dust, which will include more large particles, is analysed. Elements such as silicon and oxygen

are major components of soil dust but could also be expected in fly-ash from power stations. Where there is doubt on the origin of these elements, simple elemental analysis is not enough and analysis of impactor size fractions and electron microscope studies may be needed.

ARE ANY TRENDS VISIBLE YET?

Although for most of the sampling sites no clear trends are yet visible for any elements, there are however a few exceptions. Figure 7 shows what appears to be a gradual reduction in Mn concentrations coupled to a gradual rise in Pb concentrations in central Port Elizabeth.

Manganese shows strong maxima in summer when the prevailing winds bring dust from the ore loading facility to the city centre. Lead shows the generally observed winter maximum due to the more stagnant air conditions. Although the trends appear to be clear there is an element of doubt due to a small move in the sampling site necessitated by the destruction by fire of the city hall which initially housed the sampler. Time will be needed to separate long term trends from the strong seasonal variation. In spite of this the observed trends are in accord with other factors. The Amounts of Mn ore handled have diminished and the harbour authorities have taken measures to reduce dust emissions. Increased traffic flow and the use of high octane petrol at sea level are probably responsible for the high and increasing lead concentrations.

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ARTICLES FOR PUBLICATION IN THE CLEAN AIR JOURNAL

The Editor would welcome articles on any aspect of activities designed to improve the atmosphere in our cities.

The Journal circulation is small so that we do not expect to receive papers on the results of original research, but we think that statements on new developments in such areas as, methods for the reduction of emissions to atmosphere which have been tried out for the first time, successful methods of keeping the public up to date on the work of the local air pollution inspectors in our cities, studies of concentrations of various pollutants in city atmospheres, and allied subjects would be read with interest by all members. Articles do not need to be long and could well appear in the form of NOTES, supplying information in a matter of 200 to 500 words.

We look forward to hearing from some of you.

Table I

Correlation between light transmission and mass on the filter paper

Light transmission 1%	Mass (μg)			
	Church Square	Pretoria West	Arcadia	Scientia
90	23	23	45	75
80	90	90	140	230
70	200	210	310	495
60	360	420	625	
50	580	760		
40	860			
30				

Table II

World Health Organization, 1979

Guidelines for Exposure Limits consistent with the Protection of Public Health**

Pollutant	Concentrations ($\mu\text{g}/\text{m}^3$)	
	24-h mean	Annual arithmetic mean
1. Smoke	100 - 150	40 - 60
2. Total suspended particulates*	150 - 230	60 - 90

* tentative guidelines

** Values for smoke as measured by OECD or British daily smoke method. Adjustments may be necessary where measurements are made by other methods.

Table III

Annual Mean Concentrations of Smoke and Carbonaceous Particulates

A. Last 3 years of survey (1978 to 1980)

B. First 3 years of survey

City or Town	Geographical Situation	Concentrations ($\mu\text{g}/\text{m}^3$)							
		Central		Industrial		Residential		Black Residential	
		A	B	A	B	A	B	A	B
Cape Town	West Coast	35	70	70	100	20	20	—	—
Port Elizabeth	East Coast	30	35	60	80	15	18	15	20
Durban	East Coast	30	80	50	100	20	20	—	—
Pinetown	East Coast	35	—	25	—	15	—	—	—
Amanzimtoti	East Coast	15	—	20	—	15	—	—	—
Richards Bay	East Coast	—	—	5	—	5	—	—	—
Pietermaritzburg	Marginal Area	50	90	45	75	35	60	—	—
Estcourt	Marginal Area	55	—	20	—	—	—	—	—
Newcastle	Marginal Area	40	—	35	—	20	—	—	—
Bloemfontein	Highveld	60	115	70	120	25	35	95	130
Kroonstad	Highveld	55	65	55	85	—	—	90	—
Welkom	Highveld	—	—	—	—	35	—	60	—
Pretoria	Highveld	75	145	75	85	40	90	—	—
Boksburg	Witwatersrand	70	20	80	—	45	45	—	—
Edenvale	Witwatersrand	30	—	55	—	40	—	—	—
Germiston	Witwatersrand	45	75	60	65	30	20	—	—
Kempton Park	Witwatersrand	25	—	40	—	—	—	—	—
Randburg	Witwatersrand	—	—	—	—	25	—	—	—
Klerksdorp	Highveld	10	10	—	—	—	—	—	—
Meyerton	Highveld	25	—	30	—	20	—	—	—
Middelburg	Highveld	30	—	25	—	20	—	—	—
Pietersburg	Highveld	5	—	15	—	—	—	—	—
Springs	Eastrand	95	125	55	65	—	—	—	—
Vanderbijlpark	Highveld	30	—	35	—	30	—	—	100
Witbank	Highveld	25	—	35	—	20	—	—	—
Nelspruit	Lowveld	15	—	25	—	15	—	—	—
Benoni *	Witwatersrand	20	35	50	70	20	—	—	—
Johannesburg *	Witwatersrand	20	115	30	140	—	—	25	50

* semi-automatic filter units

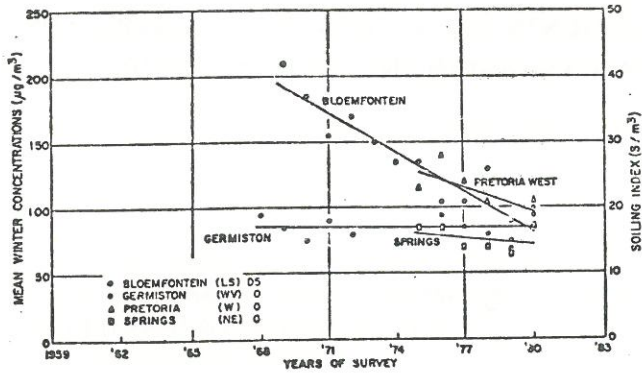


FIGURE 1

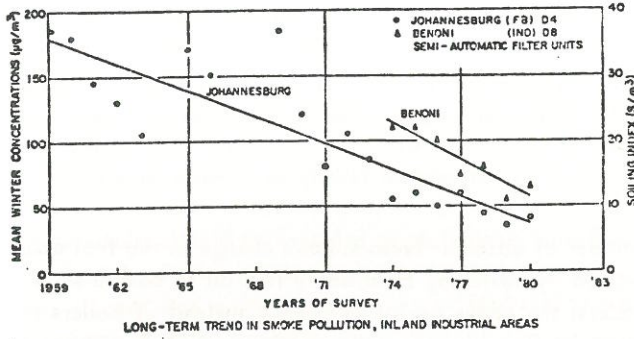


FIGURE 1

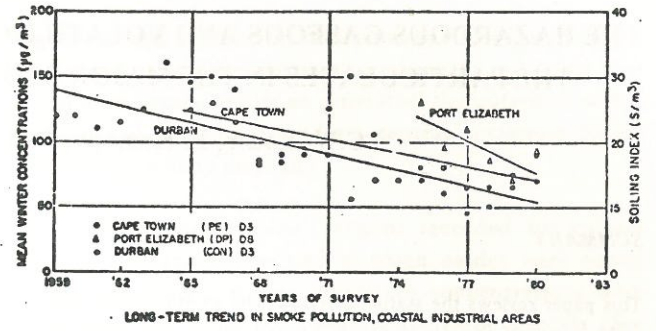


FIGURE 2

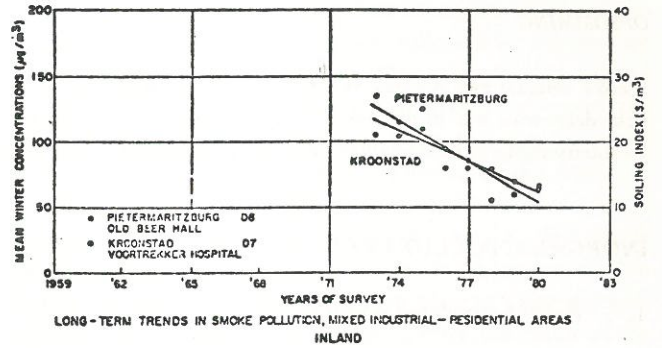


FIGURE 3

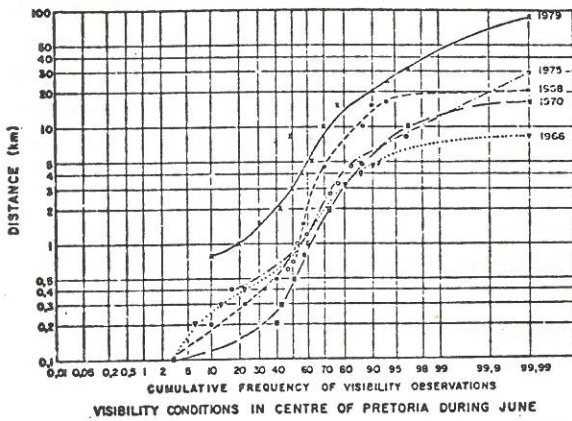


FIGURE 4

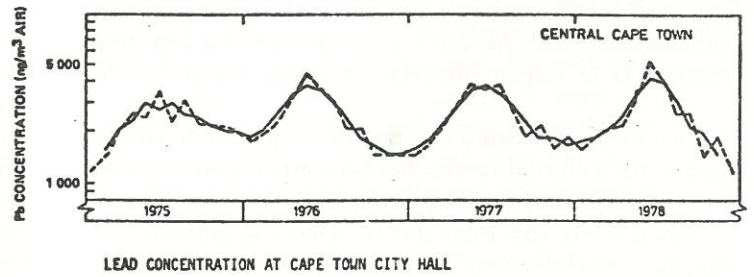


FIGURE 6

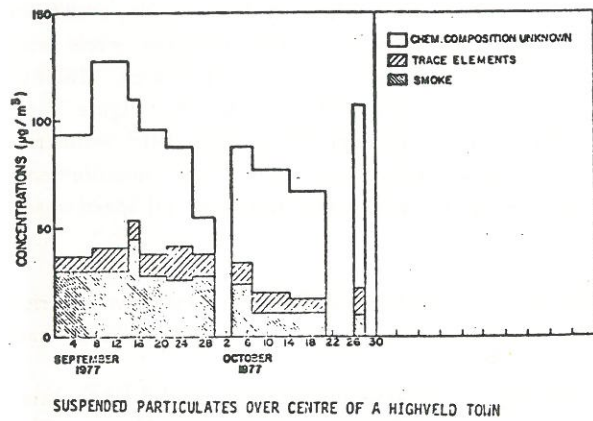
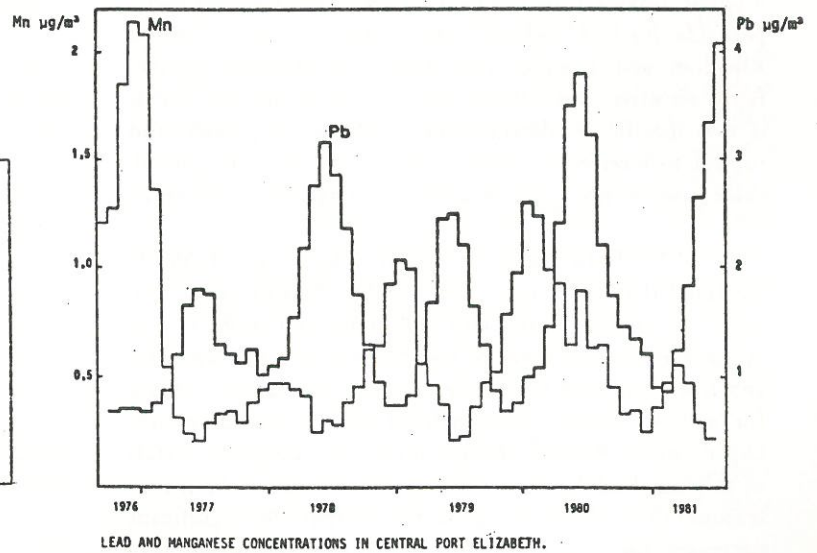


FIGURE 5



LEAD AND MANGANESE CONCENTRATIONS IN CENTRAL PORT ELIZABETH.

FIGURE 7