COMPOSITION OF SUSPENDED PARTICLES IN THE EASTERN TRANSVAAL

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ABSTRACT

The Eastern Transvaal Highveld has a large portion of industries that contribute to air pollution in South Africa. The extent of the air pollution away from the source region has not previously been studied in great detail. A one year monitoring project was undertaken in the Eastern Transvaal during 1993. The overall aims of this research were, first, to quantify the extent of particulate air pollution at remote sites on the Highveld, Escarpment and Lowveld. Second to investigate the transportation of pollutants across the Escarpment from the Highveld to the Lowveld. Samplers were commissioned at three sites in each of the aforementioned areas. Time resolved particulate samples were collected at each site and analysed by Proton Induced X-ray Emissions (PIXE). Elemental concentrations obtained from the analysis were apportioned to six source categories, namely, anthropogenic sulphur, crustal, marine, biomass burning, smelting and other. An attempt has been made in this paper to summarise the important findings of the particulate composition and concentrations at Elandsfontein, Misty Mountain and Ulusaba for three seasons of the year. The total detected particulate matter (> 10 μm) has also been compared with the synoptic scale circulation's at the same time. Particulate concentrations were found to be closely linked to the prevailing synoptic scale circulations. Accumulation of aerosols was found to occur under continental anticyclonic flow, while the atmosphere was flushed clean of pollutants with the passing of westerly and easterly troughs.

1. INTRODUCTION

Air pollution has long been the focus of much research in the Eastern Transvaal Highveld as a result of the concentration of industry in this region. As South Africa is likely to experience increased industrialisation in the future, a conscious effort needs to be made to monitor the impacts of increased air pollution on the natural environment in the surrounding and adjacent regions. Although a good understanding of the controlling mechanisms of air pollution exists in South Africa, few qualitative data are available on the extent of air pollution and on the contributions of specific sources or source types to the regional scale air quality and environmental decay. During 1992 an international research project, Southern African Fire-Atmosphere Research Initiative (SAFARI'92) was undertaken in South Africa and neighbouring countries. During SAFARI'92 the characteristics of the remote atmosphere over South Africa was studied in detail. Atmospheric chemistry data results from SAFARI'92, specifically concentrations and sources of particulate matter, highlighted the need for further studies in the Eastern Transvaal Lowveld and Escarpment regions.

The purpose of this paper is to present a summary of results from a study of the atmospheric particulate concentration and chemical composition during 1993 in the Eastern Transvaal. Time resolved ambient particle data from three sites, namely, Elandsfontein, Misty Mountain and Ulusaba Game Lodge (Ulusaba) (figure 1) will be considered in detail. The results of the source apportionment of elemental concentrations obtained from Proton induced X-ray Emissions analysis (PIXE) are presented. Total detected particulate concentrations are considered in relation to the prevailing synoptic scale circulation's at the time the samples were collected.

2. STUDY DESIGN AND SAMPLING METHODOLOGY

The sampling sites were established along the line of proposed air flow over the Eastern Transvaal (figure 2). The first site, Elandsfontein (26°15'S 29°25'E), is situated in the middle of the Eastern Transvaal Highveld 25 km south of the Witbank/Middleburg industrial region. Elandsfontein is surrounded by gently undulating rural farmland. The site was chosen so as to obtain samples that reflected the integrated urban and industrial plumes of both the Witbank/Middleburg and Gauteng regions. Misty Mountain is approximately midway between Lydenburg and Sabie on the Long Tom Pass. It is located on an exposed hill top elevated 1777 m asl. The need for an assessment of the possible impacts of Highveld pollution on the sensitive indigenous and commercial forests of the Eastern Drakensburg provided the rationale for the selection of Misty Mountain. The site was also ideally positioned to trace the movement of Highveld air masses across the Escarpment to the Lowveld through direct advection by regional scale winds. Ulusaba (420 m asl) is representative of a remote area in the Eastern Transvaal Lowveld. The sampler was situated on a small conical hill, 105 m above the otherwise flat terrain. No industrial sources occur within 80 km of Ulusaba, and any industrial material detected at this site must originate from one of three industrial source regions, viz. Nelspruit and Phalaborwa urban complexes, or the Eastern Transvaal Highveld (Piketh, 1995).

3. TIME RESOLVED PARTICLE SAMPLING

Ambient air particle samples were collected with one stage PIXE International Inc circular streaker units with a 10 μm aerodynamic diameter size selective inlet impactor (Annegarn *et al.*, 1992 and Piketh, 1995). The streaker sample consisted of a 0.4 μm pore size Nucleopore filter membrane mounted on a circular frame. The frame was attached to a motor that allowed it to rotate at one revolution per month. The filter membrane passed over 1 x 7 mm sucking orifice, through which ambient air was being drawn at 1ℓ/min. A streak of atmospheric particulate matter is left behind on the filter. Streaker frames were exposed for 672 hours or 28 days. The average volume of flow for a given period was estimated from the elapsed number of hours and the total volume of air recorded.

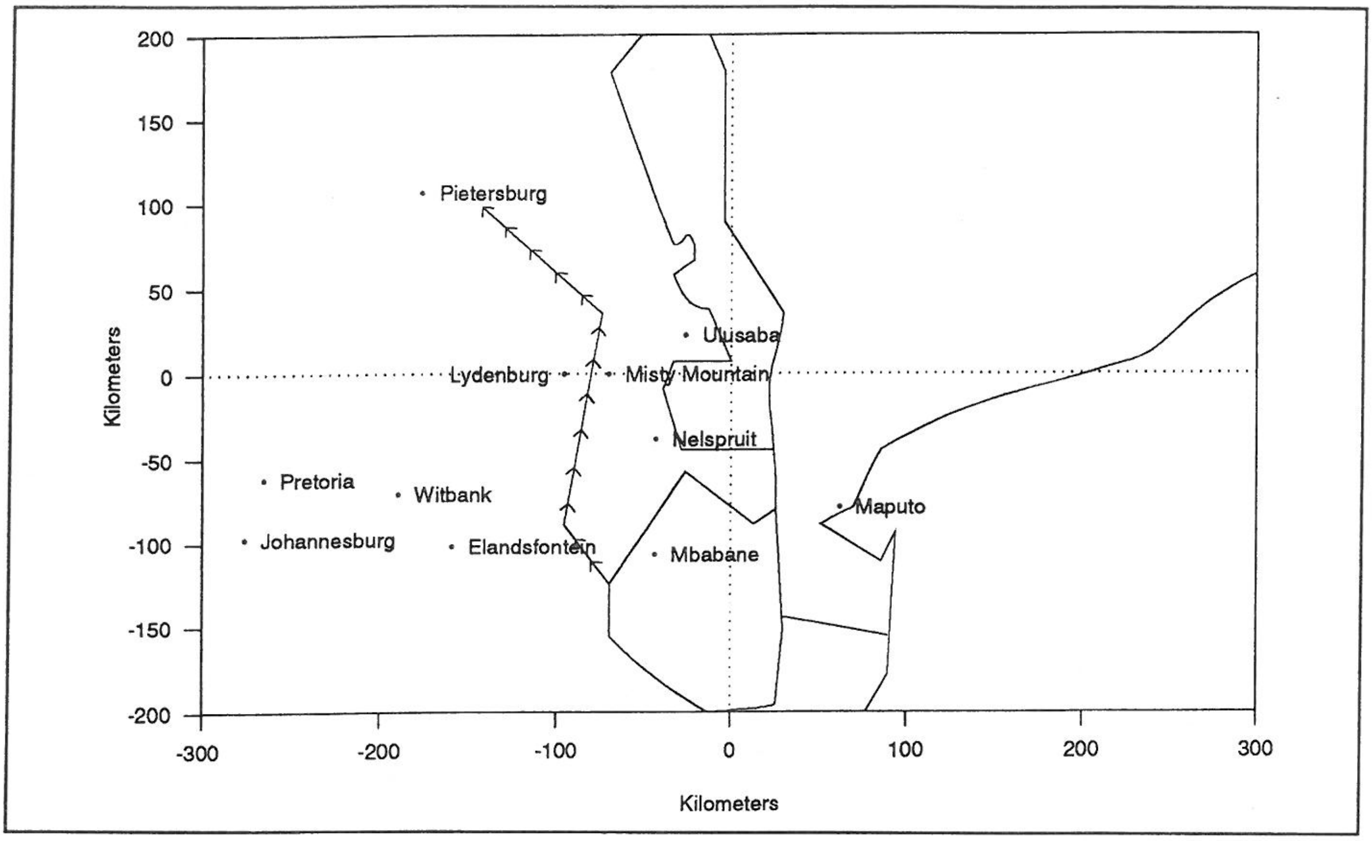

Figure 1. Location of Elandsfontein, Misty Mountain and Ulusaba in the Eastern Transvaal.

4. FILTER ANALYSIS

The exposed streaker filters were analysed by Proton Induced X-ray emissions (PIXE) with 3.2 MeV protons from the En Tandem Van de Graaff accelerator at the University of the Witwatersrand. The streak of particles was analysed in 144 sequential steps of 1 mm Each step was the equivalent of 4,5 hours of sampling. Elements analysed for and routinely observed above detection limits, included: Al, Si, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Br and Pb. Detection limits were calculated for every element in each spectra as part of the data validation procedure. Only values recorded above the detection limits were retained for data interpretation. Unexposed sections of the filters were analysed as field blanks and elemental concentrations were corrected if necessary. Elemental concentrations were calculated in $\mu g/m^3$ for each time step.

5. IDENTIFICATION AND QUANTIFICATION OF SOURCES

Source inventories are an essential part of any air quality/pollution study. A primary source inventory of the Eastern Transvaal was established in order to identify all possible contributing sources, both natural and anthropogenic, to the detected atmospheric constituents. Since only sources that would impact at regional scale were important for this study, only large scheduled industries were included in the inventory. Ten anthropogenic source categories were identified, namely, power stations, timber and related industries, metal smelters, petrochemical plants, brick and stone works, gold and coal mines, fertiliser and chemical producers, explosives manufacturers, charcoal producers and finally a general category which included all additional industrial sources. Non-industrial sources of airborne particulate matter are also important contributors to the air chemistry. For the purposes of this research crustal, marine, biomass burning (domestic wood burning included), and automobile emissions were regarded as being the most important non-industrial sources.

From the filter analysis elemental concentrations for each time step were obtained in mg/m^3. Unique elemental ratios were then used to apportion the elements to the contributing sources. Source categories were identified from a knowledge of possible sources and the elemental characteristics of their emissions.

Twelve samples were selected for analysis during the sampling campaign. The selected samples represented three seasons of 1993, summer, winter and spring. Six source categories were identified from the elemental data, anthropogenic sulphur, crustal, marine or sea salt, biomass burning, smelting and other. Anthropogenic sulphur, crustal and sea salt were the prominent sources of the particulate matter measured at Ulusaba and Misty Mountain. At Elandsfontein no sea salt emissions were detected. Table 1 summarises the contributions from each source component to the aerosol at each site. Each source category is discussed in terms of their average annual contribution and the nature of the source at each site.

Table 1. Average annual source contribution (%) of anthropogenic sulphur, crustal, sea salt, biomass burning and smelting components at Elandsfontein, Misty Mountain and Ulusaba.

	Elandsfontein	Misty Mountain	Ulusaba
Anthro sulphur	21	58	20
Crustal	69	58	49
Sea salt	0	6	23
Biomass burning	2.3	3.5	4
Smelting	2.1	0.7	0.3

5.1 Anthropogenic Sulphur

Particulate sulphur (present as SO_4) is the major secondary pollution species derived from fossil fuels. It is thus possible to estimate from this component the extent to which power stations on the Highveld for example, contribute to the degradation of the air quality in the adjacent regions. Due to the importance of this concentration ranges were defined.

Low:	[S]	<0.2 *ug*/m^3	Typical of a remote marine aerosol
Medium:	0.4> [S]	>0.2 *ug*/m^3	Typical of a remote continental aerosol
High	2.0> [S]	>0.4 *ug*/m^3	Typical of a urban/ industrial aerosol
Extreme:	[S]	>2.0 *ug*/m^3	Typical of a heavily polluted near source aerosol

Anthropogenic sulphur was consistently measured as a major contributor to the air pollution at all three sites (values ranged from 13% to 36%). Extreme episodes of sulphur concentrations occurred twice at Elandsfontein, during May and late September. For the rest of the samples taken at this site the average concentrations fell within the high range. As Elandsfontein is situated in the heart of the Highveld this was not unexpected. Average concentrations recorded at Misty Mountain and Ulusaba fall within the expected limits of a typical remote continental aerosol in two of the three sampled seasons (summer and spring). During June (winter) sulphur concentrations were consistently in the high range. High winter values would have resulted from increased fossil fuel usage during the colder months and the poorer dispersion characteristics at this time of the year. It was clear from the samples that anthropogenic produced sulphur was a significant air pollutant at a regional scale.

5.2 Crustal or Soil Emissions

The crustal component was generally the largest contributor to the ambient aerosol. This source contributes mainly coarse particles (>2.5 μm) to the suspended particles. Coarse particles are only transported short distances before they are again deposited, due mainly to gravitational settling. Dust particles only usually impact on the local environments, the source thus being tens of kilometres from the sampler. The soil concentrations showed strong reasonal variations, the highest percentage (77%) of the total aerosol being present during June. The seasonality of the crustal dust is related to the availability of unvegetated soil and thus to the amount of rainfall. The lowest contribution of soil dust (by 20% of all calculated contributions) to the atmospheric particulate concentrations occurred during February at Ulusaba. These low crustal levels could be ascribed to the rainfall in the region during this month (about 90 mm during seven events) and good vegetation ground cover in the immediate surroundings (limited potential for wind blown dust).

5.3 Marine

Marine related particulates emitted by the ocean through the stress of wind acting on water, or through the bursting of air bubbles at the surface, impacted with the greatest intensity during summer at Ulusaba (54%) The marine contribution at Ulusaba throughout the year was at least a factor of four greater than at the other sites. Marine particulates also frequently reached Misty Mountain in a diluted form, contributing a maximum of 16%, also in the February. Excess chlorine (trace element for the marine component) detected on the Highveld, coincided with other heavy metal elements and was therefore not related to the marine component.

5.4 Biomass Burning

Biomass burning was detected in the filters by excess potassium in the collected samples. Potassium was found to form about 3,8% of the total emissions from biomass burning in savannah regions of southern Africa (Salma *et al.*, 1995). This needs to be kept in mind when the biomass burning component of the pollution is considered. The biomass burning component of the aerosol showed a strong seasonal variation. Highest concentrations of biomass burning related elements were detected at all three site in early spring (September). The highest contribution as a percentage of the total detected inorganic aerosol occurred at Ulusaba in September 1993 (10%). At the Misty Mountain site the biomass burning contribution was also highest during spring. An interesting finding was that, relative to the rest of the sites, the impact of biomass burning at Misty Mountain was high throughout the year. The biomass burning component at Misty Mountain never dropped below 1% of the total detected aerosol. This was probably due to the widespread burning of slash and fire breaks by the forestry industry in the Escarpment area. Numerous tepee burners are situated on the Eastern Transvaal Escarpment in which the waste timber, bark and other unwanted vegetation is disposed of through burning. The highest concentrations of biomass burning occurred on the Highveld during September. Little prescribed burning as a result of low vegetation cover occurs on the Highveld except for the burning of the previous year's crops by farmers in the region. The high biomass burning concentrations probably resulted from burn particulates being imported from the surrounding regions, especially the north. The low contribution of the burn component to the detected particulates results from the higher pollution concentrations on the Highveld, thus making the burn component a much smaller contributor. The biomass burning concentrations are highest at Elandsfontein throughout the year.

5.5 Smelting

Smelting sources contributed a relatively small amount of particulate pollution during the sampling period. The highest contribution of smelting to the total detected particulate concentration was 1.18%. Numerous smelting industries are situated in the Witbank/Middleburg area, thus the detected smelting contribution give a clear source signature for this region. Misty Mountain consistently recorded higher contributions from a smelting source than Ulusaba. The concentrations clearly decreased over the Lowveld. Smelter plants in the Escarpment (Lydenburg) and in the Lowveld (Nelspruit) were discounted as being potential contributors, as their emissions characteristics were different to those detected at Misty Mountain and Ulusaba. The detected smelting elements at the two remote sites provided the only real evidence of direct advection of pollutants from the Highveld to the Lowveld.

6. OBSERVED SYNOPTIC CIRCULATION

One of the most important determining factors of any air pollution concentrations is the meteorological conditions that prevail at the time of sampling. Large scale synoptic circulations control the local effects and are therefore discussed below for each season during which samples were analysed. Five synoptic scale circulation patterns influence the southern African sub-continent, namely, continental anticyclones, ridging anticyclones, easterly troughs, westerly troughs and cut-off lows (Tyson *et al.*, 1988). The effects of the synoptic circulation on the total detected particulate load will also be considered.

6.1 Summer

The synoptic conditions that prevailed over the study region during February were typical of the month and generally representative of the summer months synoptic circulation. Throughout the month there was an oscillation between the high pressure system and the easterly trough/wave. The high percentage of ridging anticyclones is consistent with the literature (Preston-Whyte and Tyson, 1988) (figure 2). Generally, the continental anticyclones, the ridging anticyclones and the easterly troughs each last for between 12 - 36 hours. On two occasions the continental anticyclones dominated for just over two days (52 hours). As a result of the transient nature of the synoptic systems there was a constant rotation of synoptic scale circulations, throughout the month.

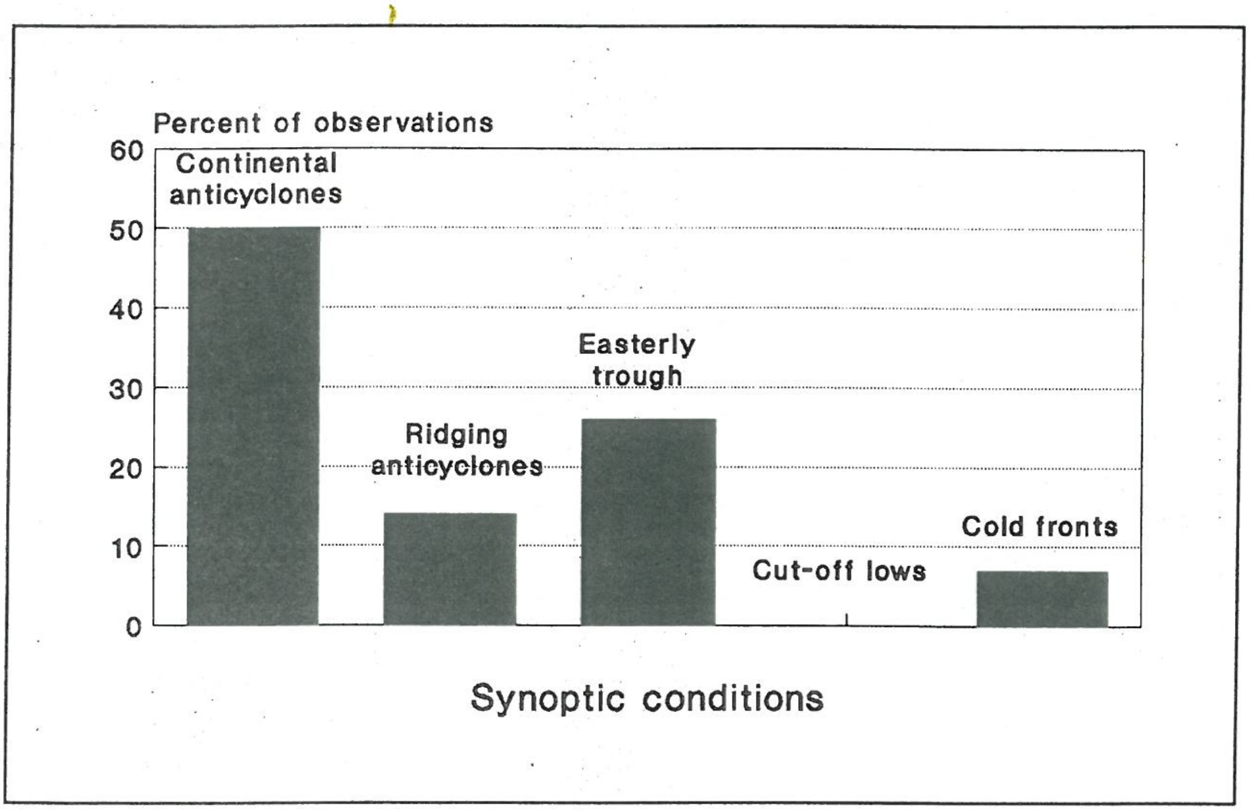

Figure 2. Synoptic scale circulation during February 1993 as a percentage of the number of observations made.

6.2 Winter

A typical winter circulation was observed during the month of June 1993. Continental anticyclonic flow prevailed over the study region for eighty percent of the month (figure 3). Westerly flow that was always associated with an advancing cold front was the only other synoptic condition that had any effect on the circulation over the subcontinent. An interesting aspect of the circulation was the fact that a continental anticyclone prevailed over the subcontinent for the last twelve days of the month. This would certainly have impacted on the concentrations of air pollutants being recirculated over the region for this extended period of time. Other circulation patterns occurred in the first two weeks of the month.

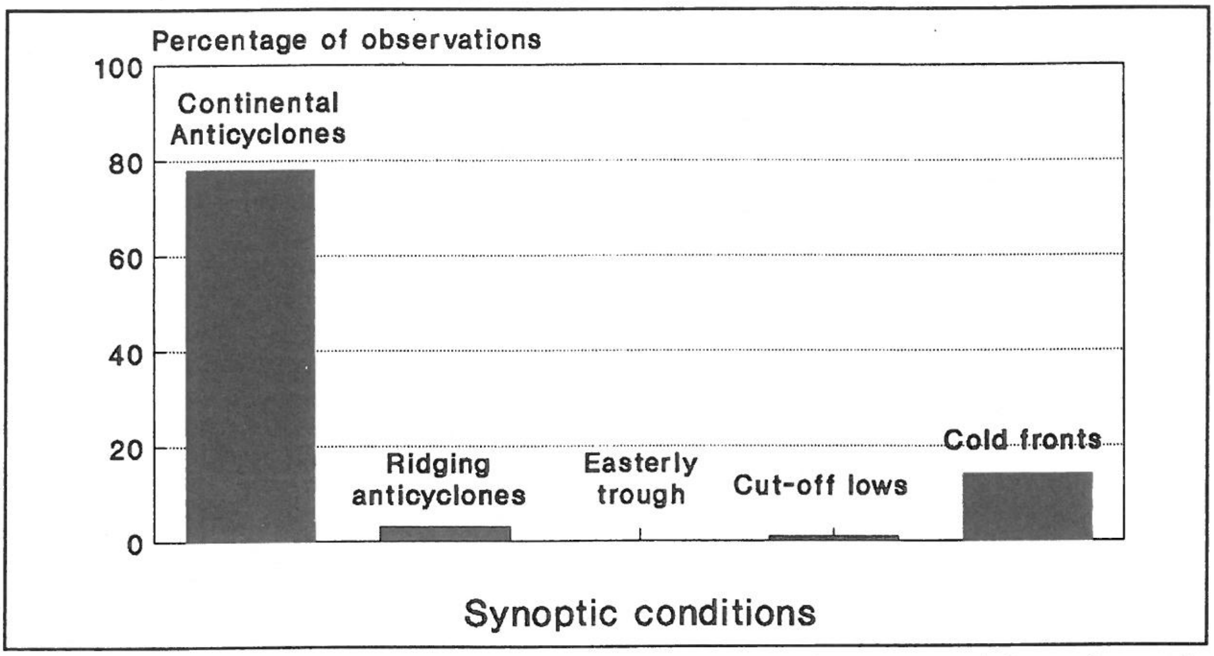

Figure 3. Synoptic scale conditions during June 1993 as a percentage of the number of observations made.

6.3 Spring

For the first four days of September continental anticyclones dominated the synoptic circulation. Spells of continental anticyclonic circulation were intermittently interrupted by short incursions of westerly waves. The westerly waves effected the circulation of the subcontinent for between 12 - 24 hours during each incursion and were then replaced by weak anticyclonic circulation. The middle and end of the month were affected by a very different circulation patterns. From the 18 - 25 September the interior of South Africa was characterised by low pressure systems (both westerly and easterly). On the 29 September a very strong easterly wave trough developed over the sub-continent and remained in place for the remainder of the month. The relative contribution of each synoptic type as a percentage of the five synoptic condition categories are given in figure 4.

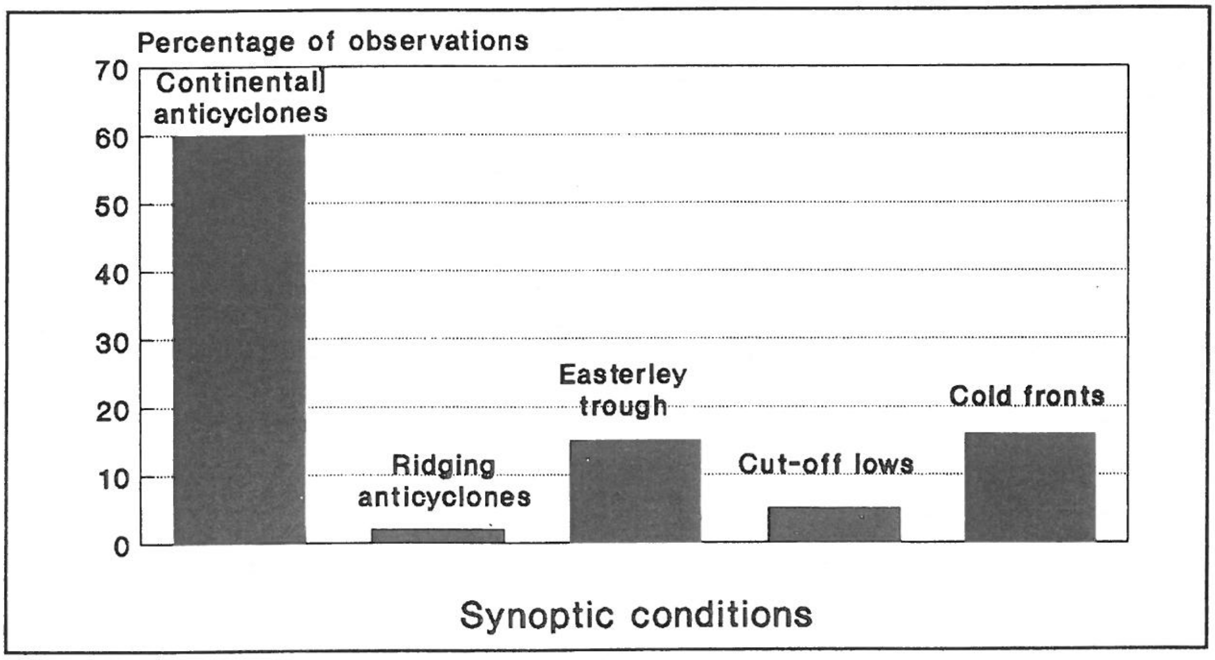

Figure 4. Synoptic scale conditions during September 1993 as a percentage of the number of observations made.

7. TOTAL PARTICULATE CONCENTRATION AND SYNOPTIC CIRCULATION

The time series plots of the total elemental concentrations for each of the analysed filters were examined in relation to the prevailing synoptic scale circulation that prevailed over the sites. General patterns could be discerned from a consideration of the two data sets. The clearest pattern was observed during February at Misty Mountain and Ulusaba. Low total particulate values were recorded in conjunction with cold front or westerly wave activity over the study region on three occasions (8, 16-17 and 22-24 February). It was true to say that during February the influence of westerly wave was always associated with cleaner air masses. However, not all clean episodes were caused by frontal systems. The same pattern of episodes was detected at the Ulusaba site.

During June the relationship between the total elemental concentrations and the synoptic scale circulation was more complex. At Elandsfontein the lowest concentrations of particulate matter were not obviously related to the synoptic circulation. On 15 June continental anticyclonic flow prevailed over the study region. The cause of the low total elemental concentrations is not clear. Westerly winds prevailed on 15 June at Elandsfontein. The low concentrations may have resulted from the a relatively clean source area to the west with wind speeds not sufficiently strong to suspend large amounts of dust particles. These low concentrations measured at Elandsfontein were not observed at Misty Mountain and Ulusaba, which confirms the local character of this event. On 6 June low total concentrations were observed at all three sampling sites. From 4 June to late on 5 June a cold front did pass over the study region followed by 18 - 20 hours of ridging anticyclonic circulation. The atmosphere was flushed out by clean maritime air, indicated by the gradual decrease of the particulate pollutants, reaching a minimum value which was maintained for the first 12 hours of resumed anticyclonic flow. From about 06h00 on 6 June the concentrations started to intensify, fluctuating about maximum values until the 11 and 12 June when a cold front again cleaned out the atmosphere.

It is clear from the above discussion that the elemental concentrations can be linked for the most part to the prevailing synoptic scale circulation. Under continental anticyclonic conditions, the total atmospheric particulate load increases until a westerly or easterly wave perturbation flushes out the atmospheric pollutants. After the onset of anticyclonic activity again, the air pollution concentrations steadily rise. This was expected to be the observed pattern. It seems, however, that extreme episodes of pollution can not be attributed to synoptic conditions. Sufficient dispersion seems to occur during the anticyclonic circulation until the atmosphere is almost completely flushed by the westerly wave systems. Extreme episodes do occur occasionally but these are a result of the development of local surface inversions during the evening during the winter months. These episodes do not persist and are estimated to last for a maximum of 6 hours (between 03h00 and 09h00 hours) (Preston-Whyte *et al.*, 1977, Tyson, *et al.*, 1976 and Preston-Whyte and Tyson, 1988). Synoptic scale circulation, specifically continental anticyclones, are however indirectly responsible for the occurrence of extreme pollution events.

8. CONCLUSIONS

Air pollution in South Africa needs to be quantified in order to evaluate the impacts that it may have on the natural environment. Levels of pollution in remote regions are able to give a good indication of uncontaminated ambient air and thus of the overall impact that anthropogenically produced pollutants have on the air quality in both industrialised and rural areas of the country. In this paper an attempt was made to summarise the extent of particulate air pollution in the Eastern Transvaal. Ambient particulate concentrations of particles below 10 μm were measured for one year (1993) at three sites, Elandsfontein, Misty Mountain and Ulusaba with a time resolution of four hours. The elemental composition of the collected samples was determined by PIXE. The detected inorganic elements could be apportioned to six contributing sources, namely, anthropogenic sulphur, soil or crustal material, marine, biomass burning, smelting and other sources.

The crustal component dominated the aerosol at the three sites for all sampling periods, showing seasonal fluctuations in the average concentrations. Anthropogenic sulphur contributed on average 10% of the detected particulate matter. Marine emissions were most prominent at Ulusaba during February. No marine related particulates were detected at Elandsfontein and excess chlorine was ascribed to a local industrial source. Smelting related elements were found in samples at all the sites. These emissions provided evidence of direct advection of pollutants across the Escarpment into the Lowveld from the Highveld. Biomass burning emissions were highest during September (spring). This was consistent with findings during the SAFARI'92 project and was expected.

Synoptic scale circulations during the February, June and September followed the expected pattern for summer, winter and spring respectively. During June continental anticyclones dominated the circulation for 80% of the time. Total detected particulate concentrations responded to the prevailing synoptic circulation. During anticyclonic flow concentrations steadily increased until the atmosphere was flushed out by either easterly or westerly trough perturbations. During winter under continental anticyclonic flow, concentration never reached excessively high levels, indicating that sufficient dilution of the pollutants was possible until the atmosphere could be cleaned out by usually a westerly wave. Extreme pollution levels are only anticipated to occur under local surface inversion conditions.

It is clear from the data presented that remote sites in South Africa are impacted on by industrial related emissions. Although the mechanisms of pollution transport are well understood for the Highveld, long range transport of pollutants over southern Africa is still poorly documented and needs to be investigated in light of the ambient chemical evidence. Impacts of the pollutants generated in industrial regions, specifically sulphur, on the vegetation of the adjacent regions also needs to be considered if natural environments of the Escarpment and Lowveld are to be preserved.

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