Assessment of Lead Exposure in Thohoyandou, South Africa

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Summary. Lead levels in different environmental media (soil, grass leaves, water, ceramics, pencil, paint, crayons and cosmetics) were determined to assess the major sources of lead exposure in Thohoyandou, South Africa. Soil and plant leaves were used as indicators of Pb pollution from vehicle exhaust emissions. After digestion with concentrated acids (HNO_3, HCl and $HClO_4$) Pb concentrations were determined in triplicate using a flame atomic absorption spectrometer. The mean Pb concentrations at the kerb of selected busy roads were 205.5 ± 90, 273.0 ± 90 and 312.8 ± 81 $\mu g\,g^{-1}$ and 154.7 ± 67, 182.9 ± 76 and 240.6 ± 66 $\mu g\,g^{-1}$ for soil and plant leaves (dry weight) respectively. These concentrations were substantially higher than the values found on soils 50 m away from the roads (97.4 ± 11 $\mu g\,g^{-1}$). Pb concentrations in plants collected further away from the road (50 m) were substantially lower (71.8 ± 9.0 $\mu g\,g^{-1}$). The observed levels on soil are lower than the UK critical value of 500 $\mu g\,g^{-1}$ for gardens and allotments; and 2000 $\mu g\,g^{-1}$ for parks and open space as well as the Canadian values for agricultural (375 $\mu g\,g^{-1}$), residential (500 $\mu g\,g^{-1}$ and industrial (1000 $\mu g\,g^{-1}$). From these data it was clear that Pb concentrations in soil samples were substantially higher than the levels obtained for plant leaves. The Pb levels in green crayons, blue crayons, pencils (from China & Germany), were 10650 ± 75.2, 8200 ± 52.4, 1160 ± 50.2, 79 ± 10.1 $\mu g\,g^{-1}$ for the inner contents; and 4870 ± 58.1, 5650 ± 55.5, 1950 ± 46.6, 60 ± 12.9 $\mu g\,g^{-1}$ for the outer surface paint respectively. The ceramics showed Pb levels of 630 ± 50.3 $\mu g\,g^{-1}$ (saucer) and 560 ± 32.2 $\mu g\,g^{-1}$ (cup), while the inner contents and outer surface paint showed 480 ± 32.4 and 318 ± 21.2 $\mu g\,g^{-1}$ of Pb respectively. Early morning tap water flush gave a Pb level of 20.6 ± 5.6 μg Pb l^{-1}. This value is higher than the WHO and FDA maximum permissible concentrations of 10 $\mu g\,l^{-1}$ and 15 $\mu g\,l^{-1}$ respectively.

Keywords: lead exposure, Thohoyandou, South Africa

Introduction

The widespread use of lead has resulted in high levels of this metal in various environmental samples. Atmospheric lead concentrations for example have been significantly increased as a result of the use of tetraethyl and tetramethyl lead as an anti-knock additive in petrol. Such elevated atmospheric Pb concentrations can pose substantial risks to humans: it has been estimated that for each 1 $\mu g\,m^{-3}$ increase in airborne lead a child's blood lead level rises between 5–6 $\mu g\,dL^{-1}$ (ATSDR, 1990). Human Pb intake can further increase due to accidental soil intake (pica behaviour of children) or due to consumption of contaminated vegetables that are in turn contaminated

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due to air pollution. Elevated blood Pb concentrations result in high blood pressure, kidney diseases, brain damage, reproductive abnormalities, coma and even death under certain circumstances (ATSDR, 1990).

Besides contaminated air, several other media can contain elevated Pb concentrations that may pose risks to humans. For example, in paint lead is present as lead chromate. Once ingested, this can be easily dissolved by gastric acid and this contributes to the bioavailability of the lead contained in the ingested dust or soil contaminated by leaded paint (Hammond, 1982). Also lead pigments are used in children's crayons and pencils and these can contribute to the total body burden of children who tend to bite pencils and therefore ingest chips and paint flakes. Measurement of lead-glazed ceramics has shown that most of the lead released comes from the glaze (Muňoz *et al.*, 1993). Some cosmetics such as eyeliners contain lead and may be a source of exposure if used on a regular basis. Lead in drinking water may come from contamination at the source, but it can also result from the water treatment and distribution system.

In South Africa, a number of studies conducted on lead exposure have concentrated on vehicle emissions as the major source (Gobler *et al.*, 1986, von Schirnding *et al.*, 1991a, 1991b; Mathee *et al.*, 2002). Consequently, studies on other sources of lead exposure within the South African environment have received little or no attention. In order to assess the local exposure of the population to lead, despite vehicle emission, other sources of lead exposure that can add to the total body burden need to be given some attention especially in those towns where no study has ever been conducted. Pencil, crayons, cosmetics and ceramics have been selected as a result of the recent influx of goods that are suspected to contain toxic metals such as lead into the South African market. These goods are sold in remote parts of the country such as Thohoyandou. It is envisaged that the results of this study will provide a baseline against which the impact of exposure to lead via different sources, particularly those mentioned above, could be measured in the future. In this paper we present evidence for the presence of lead in different environmental media commonly found in South Africa. The present study is part of an ongoing integrated environmental management programme within Thohoyandou area.

Materials and methods

Sampling sites

Three major roads were selected for soil and plant leaf sampling according to traffic densities as well as convenience of sample collection. The sampled locations are shown in Fig 1. Plant and soil samples were taken between July and August 2000.

Sample collection

Soil material   Soil samples, 30 (2–3 individual sub-samples) in total, were collected at 5 m intervals (1, 5 and 10 m) from the kerb within a depth of 1 cm, where there were no intervening pavements. Extraneous substances such as dead leaves and stones were removed from the soil. Approximately 100 g of soil was removed from each sample location using a clean stainless steel trowel. The samples for each sampling location were combined to give a composite sample mixture. These were placed in plastic bags and stored at room temperature in the laboratory until ready for use. Adequate care was taken during sampling to avoid cross contamination of samples. Soil samples collected at distances over 50 m from the roads were used to determine the extent of Pb deposition further away from the roads.

Plant material

Grass leaves were collected from 10 grass plants by cutting with a clean stainless steel pair of scissors. About 100 g grass leaves (fresh weight) consisting of a single species of grass, *Themeda Trianda*, were collected at a height of 1 m above ground at a distance of 5 m from the kerb. The grass leaves were placed in plastic bags, taken to the laboratory, rinsed under running deionised water from a 50 l plastic container and then stored at room temperature in the laboratory until further use. Grass leaves of the same species were also collected from distances over 50 m as indicated above.

Ceramic ware, crayon, pencil and eyeliner samples

Different ceramic ware, crayons, pencils and eyeliner samples were randomly selected from products available in the shops in Thohoyandou. Paint flakes
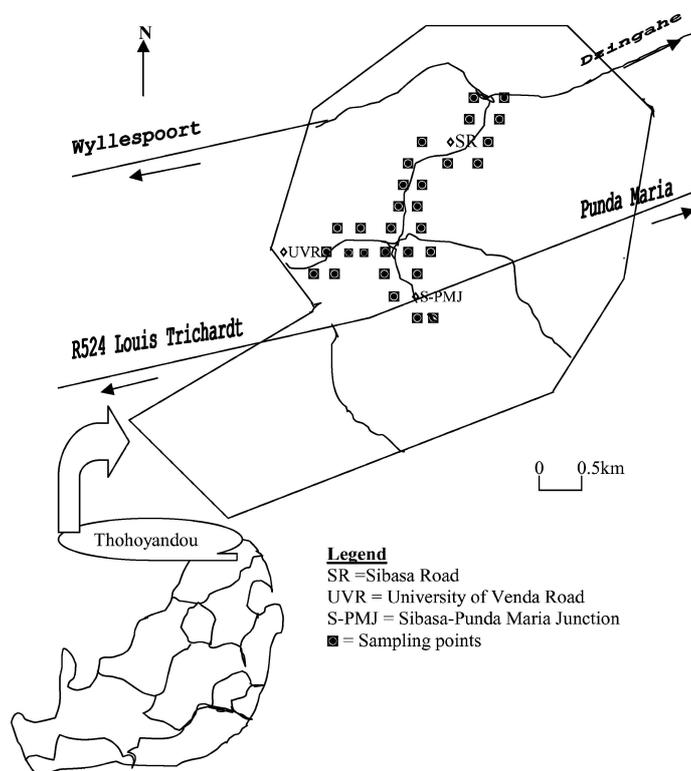
Figure 1. A sketch map of South Africa (bottom left) and Thohoyandou (top right) showing the sampling points.

were collected from buildings within the University of Venda using stainless steel materials. The material was stored in plastic bags and taken to the laboratory.

Water samples

About 100 ml of water samples were randomly collected from households and from the University of Venda student hostels using lead-free plastic containers that had been immersed for several hours in 5% HNO_3 and were thoroughly rinsed with deionised water.

Determination of Pb in soil materials

Soil samples were first dried at room temperature, sieved through a 150 μm sieve and finally ground using a clean mortar and pestle. 2 g of the sieved, ground and oven dried (110°C for 4 h) sample was placed into a 100 ml beaker to which concentrated HNO_3 (3.0 ml) and concentrated HCl (8.0 ml) were added. The flask was covered and allowed to digest for 4 h. The flask

was then heated and maintained at 180°C. Additional digestion solution was added as necessary. The solution was allowed to cool; consequently the solution was filtered through a Whatman 48 filter paper into a 50 ml volumetric flask and diluted to the mark with 2 M HNO_3. Soil samples taken from distances beyond 50 m, were subjected to the same digestion treatment. A blank was prepared as described above but without the sample.

Determination of Pb in plant material

The collected plant biomass (100 g grass leaves fresh weight) was dried at room temperature and placed in a thermostat oven at 110°C for 2 hours. 20 g of the dried material was repeatedly ground to uniform size using a mortar. About 1–2 g of the ground sample was weighed into a beaker and 20 ml of a 6:1 mixture of HNO_3 and $HClO_4$ (70%) were added. The mixture was gently heated in a fume-cupboard to 70–80°C until most of the samples had dissolved. The temperature was then raised to 140–150°C to ensure complete dissolution.

Consequently, the solution left in the beaker (8–10 ml) was filtered into a 50 ml volumetric flask through a Whatman 48 filter paper and made up to the mark with 2 M HNO_3. Grass leaves collected at distances over 50 m were subjected to the same treatment. A blank was prepared as well.

Determination of Pb in ceramic ware and paint flakes

Samples of ceramic ware and paint flakes were separately crushed in a mortar until they were in fine particles. 25 g of each sample was transferred into silicon crucibles and placed into a muffle furnace set at a temperature of 600°C for 1 h. Consequently, the samples were removed and allowed to cool in a desiccator. About 2 g of each sample was placed into a beaker and 15 ml HNO_3 added to each sample, and then warmed gently on a hot plate for 2 h. Additional digestion solution was added as necessary. The solution left in the beaker was allowed to cool, filtered through a Whatman 48 filter paper into a 50 ml volumetric flask and made up to the mark with 2 M HNO_3.

Determination of Pb in the inner and outer contents of pencil, crayon and eyeliner

The paints on the outer surfaces of the pencil, crayon and eyeliner were carefully removed using a stainless steel scalpel. The material was crushed into a fine powder using a mortar. The inside contents of the pencils, crayons and eyeliners were subjected to the same treatment as described above. The ground outer and the inside contents of the pencil, crayon and eyeliner were placed into a muffle furnace set at 600°C and then treated as described above.

Determination of Pb in drinking water

100 ml of tap water (early morning and afternoon) collected randomly from different households and the student hostels were digested using the open beaker acid digestion method (DWAF, 1992). The addition of HNO_3 followed by gentle heating ensures the concentration of the analyte in the sample. In the present study, 5 ml HNO_3 was added into the water sample in a beaker. Each flask was placed on a hot plate and heated very gently for about 2 h. More HNO_3 solution was added in order to ensure adequate digestion. The samples were allowed to cool and were then filtered using a Whatman 48 filter paper and made up to the mark as described earlier. Blanks were prepared as well.

Pb determination

A Varian Spectra AA 220 atomic absorption spectrophotometer equipped with a single slot burner was used. The analytical signals were recorded using a program for data handling controlled by a microprocessor. All used glassware was of pyrex brand. The material was soaked in dilute HNO_3 for a day, and thoroughly rinsed several times with double distilled deionised water, and oven dried before use. All reagents used in the present study were of analytical grade. Lead standard solutions were prepared from a stock lead standard solution (Fisher Scientific) with a concentration of 1000 $\mu g\ g^{-1}$. Double distilled deionised water was used for preparing the standard solutions.

The detection limit of the FAAS was determined by using the lowest concentration of Pb standard that gave the least signal with the flame AAS. Six replicates were prepared and analyzed. The detection limit was then determined from 3δ (δ = standard deviation of the replicate determination (Miler and Miler, 1998)

Statistical analysis

All samples were prepared in triplicate and then analyzed using the Varian Spectra AA220 flame atomic absorption spectrometer. From the triplicate measurements, the mean Pb concentrations were calculated and expressed as $\mu g\ g^{-1}$ and $\mu g\ l^{-1}$ for the solid and water samples respectively.

The mean and standard deviations were calculated from the measurements and statistical differences determined by Wilcoxon test at 95%.

Due to unavailability of reference samples in our laboratory, a quality control check had to be conducted by spiking undigested samples (1–1.5 g) soil, grass leaves, ceramic ware, crayon, eyeliners and pencils, prepared as described above, with known concentration of Pb standard solutions. In the case of water, doubly distilled and deionized water was spiked. From the measurements, the percentage recoveries and the standard deviations were calculated.

Results and discussion

Table 1 shows the quality control test conducted on soil and grass leaf samples in order to verify the efficiency of sample treatment procedure. The high percentage recovery shown in Table 1, validated the sample treatment procedure used in the present study. The minimum detection limit was found to be 0.085 μg l^{-1}.

The mean Pb concentrations of soil and grass leaf samples obtained for the roads at a distance of 5 m are shown in Table 2. The highest Pb soil is found

Table 1. Quality control test

Sample	Absorbance of sample without Pb standard A	Absorbance of Pb standard B	Absorbance of sample with added Pb standard C	% recovery
UV soil[a]	0.179	0.246	0.418	97.2
SR soil[b]	0.194	0.246	0.424	93.5
S-PMJ soil[c]	0.272	0.246	0.506	95.1
UV leaves[d]	0.098	0.246	0.325	92.3
SR leaves[e]	0.158	0.246	0.401	98.7
S-PMJ leaves[f]	0.201	0.246	0.435	95.1
Ceramic ware				
Saucer	0.180	0.246	0.385	83.3
Cup	0.210	0.246	0.420	85.4
Pencil				
Grey	0.150	0.246	0.378	92.7
Crayon				
Blue	0.075	0.246	0.300	91.5
Green	0.169	0.246	0.405	96.0
Paint flake	0.205	0.246	0.428	90.7
Tap water	0.005	0.246	0.240	95.5

[a]University of Venda Road soil.
[b]Sibasa Road soil.
[c]Sibasa-Punda Maria Junction soil.
[d]University of Venda Road leaves.
[e]Sibasa Road leaves.
[f]Sibasa-Punda Maria Junction leaves.

Table 2. Lead concentrations in soil and grass leaf samples along selected roads in Thohoyandou

Site	Mean Pb concentration (μg g^{-1}) $\pm$ SD		($\alpha = 0.05$)
	Soil (n = 12)	Leaves (DW) (n = 10)	
UVR	205.5 $\pm$ 90	154.7 $\pm$ 67	Significant
SR	273.0 $\pm$ 90	182.9 $\pm$ 76	Significant
SPMJ	312.8 $\pm$ 81	240.6 $\pm$ 66	Significant
Distance >50	97.4 $\pm$ 11	71.8 $\pm$ 9.0	Not significant

SD = standard deviation; DW = dry weight; UVR = University of Venda Road; SR = Sibasa Road; SPMJ = Sibasa Punda-Maria Junction.

in the Sibasa-Punda Maria Junction sample. This is because the junction experiences more than 50% of the traffic in and out of Thohoyandou. The second busiest road, Sibasa Road, shows the second highest Pb value of 273.0 $\pm$ 90 μg g^{-1}. The University of Venda Road that experiences low traffic, mainly from the university shows the lowest value, 205.5 $\pm$ 90 μg g^{-1} Pb. This trend is repeated for grass leaf samples with the University of Venda Road showing the lowest Pb concentration of 154.7 $\pm$ 67 μg g^{-1}. This value is followed by a Pb concentration of 182.9 $\pm$ 76 μg g^{-1} shown by Sibasa Road. The grass leaf samples obtained from Sibasa-Punda Maria Junction show the highest Pb concentration (240.6 $\pm$ 66 μg g^{-1}). Table 2 clearly shows that the Pb levels for soil and grass leaves are higher than the values (97.4 $\pm$ 11 and 71.8 $\pm$ 9.0 μg g^{-1}) for soil and grass leaves respectively obtained 50 m away from the road. The observed difference suggests some level of Pb pollution probably from vehicle emissions since there are no high level industrial activities within the area. The Pb concentration obtained for distances greater than 50 m for soil (97.4 $\pm$ 11 μg g^{-1} Pb) and grass leaves (71.8 $\pm$ 9.0 μg g^{-1} Pb) in the present study substantially exceeds reported background values of 25 μg g^{-1} Pb in soil and 5–10 μg g^{-1} Pb in vegetation respectively (UK Trigger concentration, 1987, Canadian Environmental Quality, 1992). One of the sources of Pb obtained in the soil samples in the present study may have come from the deposition of Pb following emission from vehicles. Roadside soils have been shown to have considerable contamination due to both direct deposition of vehicle-derived metal and to relocation of metals deposited on the road surface (Harrison *et al.*, 1981). Metals such as Pb, once deposited in soil are usually strongly bound to the soil humic and clay colloids; therefore Pb in general has low bioavailability. However, this low bioavailability can be altered at low pH, since it is well known that nearly all metals are most soluble and bioavailable at low pH. Consequently, toxicity problems are likely to be more severe in acid environment. Where sulphide pollution is a problem, the weathering of sulphide that tends to increase the acidity of the soil can exacerbate the situation. As far as the leaves are concerned, the major route of contamination is known to be through foliar absorption of atmospheric deposits on the leaves (Kloke, 1984). The role of root uptake may not have been significant since the transfer coefficient of Pb is relatively low, 0.01–0.1 (Kloke, 1984).

Table 3. Lead concentration in soil samples in Thohoyandou Town according to distance ($n = 9$)

Site/Distance from the road (m)	Mean Pb concentration (μg g^{-1}) $\pm$ SD		
	1	5	10
UVR	234.4 $\pm$ 94	205.5 $\pm$ 90	200.9 $\pm$ 51
SR	280.1 $\pm$ 101	273.0 $\pm$ 90	264.6 $\pm$ 75
SPMJ	317.7 $\pm$ 146	312.8 $\pm$ 81.	317.6 $\pm$ 25

SD = Standard deviation.

The lead concentration in soil samples according to distance is shown in Table 3. In general Pb concentration tend to decrease with distance. This decrease, however, is not significant; moreover, for Sibasa-Punda Maria Junction a slight increase is found. The slight increase observed for Sibasa-Punda Maria Junction, may have been influenced by the slight slope existing at this junction as was observed during sampling. During rain, water would run down the slope, carrying with it soil and other particulate matter from the higher elevation and depositing these and the accompanying Pb on the lower part. Similar result on the impact of slope on Pb particle distribution in soil, has also been reported by Mmbaga and Semu (1999).

The UK critical values for soil Pb for the reclamation of contaminated land are given as 500 μg g^{-1} for gardens and allotments; and 2000 μg g^{-1} for parks, playing fields and open space (UK Trigger concentration, 1987). These values are substantially higher than the values obtained for soil in the present study. Also compared to the Canadian National Classification System for Pb contaminated agricultural (375 μg g^{-1}), residential (500 μg g^{-1}) and industrial (1000 μg g^{-1}) soils, the values obtained in the selected roads in Thohoyandou, are lower than these values (Canadian Environmental quality, 1992). However, the results obtained in this study for soil samples fall in the range of 50–300 μg g^{-1}, which is a range for Pb concentrations in urban soils and dusts in African towns and cities (Sedman, 1989, Barnes, 1990, Van Wijnen *et al.*, 1990). What is worrisome is that factors such as vibrant outdoor lifestyle, the endemic dusty environment, poor nutrition and health and poor hygiene practices, all of which are prevalent in Thohoyandou, can combine to increase the level of exposure. These determinants have been shown to strongly influence metal exposure and absorption; and where soil Pb concentration is increased by 1000 μg g^{-1}, the geometric mean blood lead (Pb-B) level can increase by 18 μg dL^{-1} (Matte

et al., 1991). The health effects of such a two-fold difference in absorption rate can be drastic.

According to Alloway and Ayre (1997), the normal and phytotoxic Pb concentrations generally found in plant leaves are 5–10 and 30–300 μg g^{-1} (fresh weight) respectively. The Pb concentrations obtained for the plant leaves in the present study are within the phytotoxicity range, although no phytotoxicity test was conducted in the present study. Foliar absorption of atmospheric Pb deposits on plant leaves may account as the major route, since for metals such as lead, the transfer coefficient is very low. The flood that occurred in the Limpopo Province, South Africa in 2000, may have contributed to the results obtained in the present study. The flood caused a landslide, carrying away roads, vegetation and bridges. It is most likely, that a good quantity of Pb particles that may have been deposited on soil in Thohoyandou town, before the flood, was washed away during this period, since sampling was conducted seven months after the flood.

The mean Pb concentrations in environmental samples from different countries are shown in Table 4. The results show a wide variety of lead contents depending on the origin as well as the sample type. The crayons and the pencils show the highest Pb values which substantially exceed the US Food and Drug

Table 4. Mean lead concentrations in environmental samples from different countries

Origin	Type of sample	No of samples	Pb concentration (μg g^{-1})
China	Crayon		
	Green	5	10650 $\pm$ 75.2 (4870 $\pm$ 58.1)
	Blue	5	8200 $\pm$ 52.4 (5650 $\pm$ 55.5)
China	Pencil		
	Grey	5	1160 $\pm$ 50.2 (1950 $\pm$ 46.5)
Germany	Pencil		
	Grey	5	79 $\pm$ 10.1 (60 $\pm$ 12.9)
China	Ceramic ware		
	Saucer	2	630 $\pm$ 48.3
	Cup	2	560 $\pm$ 32.5
India	Eyeliner	5	480 $\pm$ 30.4 (750 $\pm$ 40.7)
South Africa	Paint flakes	5	318 $\pm$ 21.2
South Africa	Tap water		
	Morning draw	10	20.6 $\pm$ 5.6 (μg l^{-1})
	Afternoon draw	10	ND

Figures in parenthesis refer to outer surface paint.
ND = Not detectable (detection limit = 0.085 μg l^{-1}).

Administration's recommended level of 90 μg g^{-1} for crayons and pencils (USFDA, 1988). This source of exposure is significant and can contribute to the total Pb body burden of children who have the common habit of chewing crayons and pencils. It can also be observed from Table 4, that the Pb concentrations in the outer surfaces of crayons from China are lower than the inner contents. This trend is however, not repeated with the outer surface paint on the grey pencil (from China) with a Pb concentration of 1950 μg g^{-1}. This value is by far lower than the values (3194 μg g^{-1}) recorded by Romieu (1994) in some Mexican pencils. The ceramics show Pb concentrations of 630 μg g^{-1} and 560 μg g^{-1} for saucer and cup respectively. These values are still higher than 500 μg g^{-1} recommended by USFDA (1988). With the observed high levels of Pb in the ceramics used in this study, it is possible that acidic substances such as chili can easily induce leaching of Pb since the solubility of Pb increases with decrease in pH value. Consequently, food items and beverages prepared in such ceramic ware, are likely to show high levels of Pb. The Pb concentrations for cosmetic (eyeliner) are 480 $\pm$ 30.4 and 750 $\pm$ 40.7 for the inner and outer contents respectively. If used on a regular basis, this can present a major source of exposure especially for women. From Table 4, the concentration of Pb in paint flakes used in the present study (318 μg g^{-1}) is substantially lower than 600 μg g^{-1}, recommended by the South African Bureau of Standards (SABS, 1999). However, this is not so with the Pb level in the early morning draw (20.6 μg l^{-1}) which substantially exceeds the acceptable level for human consumption, 10.0 μg l^{-1}, by SABS (1999) and the WHO (1996). The observed Pb level may be attributed to the slight acidity of the water (pH 6.3–6.4) as well as the use of Pb pipes in some parts of Thohoyandou. Fergusson (1990) also found elevated Pb concentration, over 50 μg l^{-1}, in first-draw drinking water in 24 towns in the UK. This was attributed to the use of Pb pipes and solder as well as the acidity of the water. The presence of lead in drinking water is undesirable as a result of its toxicity. Concentrations below 50 μg l^{-1} can produce a detectable elevation of blood lead and this gives rise to concern about the possibility of consequential problems. Of particular importance is the possibility of neuro-physiological effects influencing learning ability and general behaviour in children, who with pregnant women and infants are considered to be the most sensitive group of the population (Packham, 1996). Concentrations greater than 50 μg l^{-1} will definitely increase the blood lead level and this can lead to several complications such as the inhibition of δ-aminolevulinic acid dehydrase whose catalytic function is essential for biosynthesis of heme. Because this impairs the production of hemoglobin, oxygen starvation and anemia may occur. It has also been suggested that blood lead levels may be associated with biochemical abnormalities in children's brain, since there is a direct relationship between the activities of blood and brain enzymes (Bellinger *et al.*, 1986; Baghurst *et al.*, 1992).

Conclusions

The present study has shown some Pb contamination of soil and grass leaf samples in Thohoyandou. Pb values for soil and plant leaves were found to be substantially higher on roads than values found at 50 m from roads. While the Pb concentration in the soil sample has been found to be lower than some internationally recommended values, there is still cause for concern considering the vibrant lifestyle, the open type of dwellings and endemic dusty environment which can add to the exposure level of the local population. The same argument can be used for the Pb concentration on plant leaves. The Pb contents in the other environmental samples analysed in the present study indicate that Pb exposure, particularly from the other sources, is still reason for concern in South Africa. The major sources are, crayons, pencils, ceramics and cosmetics (eyeliner). The results presented in this study emphasize the need for strict regulatory policies on the importation of commodities into South Africa, in order to protect the population from Pb exposure.

Acknowledgments

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