

Arsenic residues in soil at cattle dip tanks in the Vhembe district, Limpopo Province, South Africa

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Arsenic-based compounds have been used for cattle dipping for about half a century to combat East Coast Fever in cattle in South Africa. The government introduced a compulsory dipping programme in communal areas to eradicate the disease in 1911. Concern has been raised regarding the ecological legacy of the use of arsenic-based compounds in these areas. We investigated the incidence of arsenic residue in soil at 10 dip sites in the Vhembe district of Limpopo Province, South Africa. We found high levels of arsenic contamination at a depth of 300 mm at some sites. Control samples indicated that these high arsenic levels are the result of the application of inorganic arsenic. Variation of arsenic concentrations is attributed to duration of exposure to the chemical, soil properties and distance from the dip tank. Concerns are raised regarding the structural condition of the dip tanks, encroaching villages and possible health threats to the human population in the area.

Introduction

We report on arsenic levels in soil around cattle dip tanks in the Vhembe district of the Limpopo Province, South Africa. Here, as was the case elsewhere in South Africa, arsenic-based dipping compounds were used for many decades to combat East Coast Fever (ECF) among cattle.

Arsenic compounds are divided into three major groups: inorganic arsenic compounds, organic arsenic compounds and arsine gas. Inorganic arsenic is more toxic than the organic variant; and arsenite (AsO_3^{3-}) is more toxic than arsenate (AsO_4^{3-}).^{1,2} Arsenic in the environment is of either geogenic or anthropogenic origin. Arsenic with a geogenic origin is usually related to background material and minerals such as antimony, copper, iron, lead, nickel and silver. Anthropogenic sources of arsenic include the mining and smelter industry, burning of coal, treatment of wood, tanneries, the pharmaceutical and glass industries, and pesticides.

Arsenic is a dangerous, persistent, non-biodegradable and accumulative substance. In the United States of America (USA) it is classified as a class A carcinogen. In humans, high concentrations cause arsenic poisoning with detrimental effects.^{2,3} Because of the intrinsic danger of arsenic to society, the World Health Organization has expressed a safe maximum permissible value for drinking water of 0.01 mg/L or parts per million.⁴ The fatal dose for humans of ingested arsenic is between 70 mg and 180 mg.¹ Human consumption of arsenic-contaminated foods over a long time may lead to arsenicosis, a chronic illness that produces skin disorders, gangrene and cancer of the kidneys and bladder.⁵ Where it accumulates in the nervous system, arsenic may induce mental-related problems. Chronic exposure to small amounts of arsenic in drinking water increases the risk of cancer and other diseases in humans.⁶

Most of the evidence on geogenic arsenic contamination relates to Asia.^{7,8} In southeast Bangladesh, arsenic contamination of water sometimes exceeds 1000 mg/L. In contrast, arsenic concentrations in uncontaminated soil in North America generally do not exceed 15 mg/kg, while in the United Kingdom, the mean arsenic concentration in rural soil is 10 mg/kg.^{2,9} In areas considered unlikely to have been exposed to anthropogenic sources of arsenic, median concentrations of 3.9 mg/kg and 0.6 mg/kg were, respectively, found at 15 sites in South Australia and at 6 sites in Tasmania.¹⁰ Arsenic concentrations in soil in rice fields in China varied between 1.29 mg/kg and 25.28 mg/kg with a mean of 6.04 mg/kg, well below the ≤ 30 mg/kg arsenic soil limits stipulated for agricultural land in that country.⁵

In areas such as Australia, New Zealand and the southern states of the USA, past livestock dipping practices are often blamed for high arsenic concentrations in soil.^{11,12} Soil samples from seven sheep dip sites in Australia and five cattle dip sites in Florida (USA) revealed arsenic concentrations between 31.3 mg/kg and 2143 mg/kg.¹³ Consequently, a number of countries have laid down guidelines for the management of arsenic-contaminated soil at dip sites. The Australian and New Zealand Environment and Conservation Council (ANZECC), for example, has determined Interim Sediment Quality Guidelines (ISQGs) for arsenic in soil with an ISQG-low of 20 mg/kg and an ISQG-high of 70 mg/kg.¹⁴ The ANZECC policy states that when the measured arsenic concentration in soil is below 20 mg/kg no action is required; when it is between 20 mg/kg and 70 mg/kg it should be assessed against background concentrations; and when the measurement exceeds either the ISQG-high or both the ISQC-low indicator and the background concentration, an assessment of bioavailability should be conducted.

East Coast Fever, cattle dipping and arsenic in South Africa

Arsenic-based animal dipping compounds were introduced in South Africa in 1893. Following the outbreak of ECF in 1901, cattle dipping became general practice in this country. Towards the end of the South African War in 1902, there was a shortage of cattle in the country. Imported cattle en route from Australia and India were sometimes offloaded for grazing at Mombasa – an ECF endemic area – before proceeding to South Africa and then Rhodesia (now Zimbabwe).^{15,16} The disease first appeared at Mutare in Rhodesia in 1901 and at Komatiport in South Africa in 1902. Thereafter, it spread rapidly along transport routes from the coast inland.¹⁷ Between 1901 and 1960, when ECF was finally eradicated in southern Africa, approximately 1.5 million cattle either died from the disease or were slaughtered to prevent the spread of infection.¹⁶

Compulsory dipping for ECF was originally introduced in terms of the *Stock Disease Act of 1911*. The former Venda area of South Africa (now the Vhembe district of Limpopo Province) is infested with the brown ear tick

(*Rhipicephalus appendiculatus*), which is a vector of *Theileria parva* – the parasite that causes ECF. The region was therefore subjected to the ECF national dipping programme, starting in 1915 when the first dip tank was erected.^{18,19} The dipping programme in communal areas (such as Venda) was administered by the Department of Native Affairs with technical services provided by the Division of Veterinary Services. Thousands of dip tanks were built and by the 1920s all affected areas had on average one tank for every thousand head of cattle. By around 1960, when ECF was finally eradicated in southern Africa, 10 million cattle were being dipped every 7–14 days.²⁰ Arsenic oxide (As₂O₃) and trioxide (As₂O₅) compounds were most commonly used in the dipping programme.²¹ Despite the eradication of ECF, compulsory cattle dipping continued in communal areas to prevent the outbreak of foot and mouth disease (FMD).

Although the use of arsenic-based animal dipping compounds was banned in 1983, the after-effects of arsenic compounds still pose a threat because of the adverse characteristics of the chemical. It is envisaged, for example, that indigenous yellow- and red-billed oxpeckers (*Buphagus africanus* and *Buphagus erythrorhynchus*, respectively) could soon become extinct in South Africa as result of arsenic poisoning.²² Concern about the impact of arsenic-based dipping compounds on human health in South Africa was expressed as early as the 1940s.²³ Present-day research on arsenic contamination in South Africa includes the search for arsenic-resistant bacterial genes in mining ash and the pollution levels in treated wood.²⁴⁻²⁷ However, research in the field of arsenic contamination of soil at dip tanks – the focus of our study – has been limited. However, one study, conducted by Moremedi and Okonkwo²⁸ at Ka-Xikundu village close to the Luvuvhu River, also in the Vhembe district, reported high arsenic levels (above > 1000 mg/kg) close to a dip site at the surface, and at 50-mm and 100-mm depths, and a significantly lower concentration of about 0.15 mg/kg at a control site some distance away. The authors recommended that the risk posed by historical arsenic-based dip operations to the immediate environment, water resources and vegetation be investigated. We aim to extend the work on arsenic contamination in soil resulting from past cattle dipping practices.

Material and methods

Study area

The Vhembe district is, apart from a few towns such as Thohoyandou, largely a rural area under communal occupation. Prior to the South African War, various skirmishes occurred between the Vhavenda and the South African Republic. After the war, the Vhavenda tribe was subjugated in 1905 by the victorious British government.^{19,29} In 1910, Venda became

part of a unified South Africa. From 1979 the area was administered as an ‘independent’ homeland before being incorporated into a re-united South Africa in 1994. For the Vhavenda, cattle are an indication of wealth. Apart from their monetary value, cattle also are used as an important currency for various social activities. The Vhavenda therefore resisted confinement to a limited territory, the forced reorganisation of their agricultural system through ‘betterment planning’, and being forced by the state to comply with compulsory cattle dipping.^{30,31} However, over time, through the introduction of incentives and the involvement of tribal chiefs, the dipping programme gathered momentum.

Distribution of sample sites

Soil samples were collected from 10 dip sites in the Vhembe district. The selection of the dip sites was based on the dates of construction of the various dip tanks, soil characteristics and ecoregions. The respective dip sites include a group of 54 tanks that were established in the study area between 1915 and 1955. The dip tanks are situated at the villages of Khubvi, Mukula, Rambuda, Sambandou, Thengwe, Tshandama, Tshifudi, Tshikuwi, Tshituni and Tshivhulani (Table 1).

The Water Research Commission classifies the Vhembe district into three ecological regions.³² In Ecoregion 2.01 (central highland area), the soil around the Khubvi and Mukula dip tanks is highly weathered and consists of compacted red clay; around the Tshifudi dip tank, sandy loam with organic matter is prevalent; and around the Tshivhulani dip tank, deep red clays predominate. In Ecoregion 5.04 (northeastern area), the soil around the Rambuda dip tank is red and loamy with a high content of organic matter; around the Sambandou tank, the soil is sandy loam with high levels of organic matter; and the Thengwe and Tshandama dip tank areas have sandy soil with little organic matter. In Ecoregion 5.03 (western area), red loam soil, heavily weathered because of compaction, is found around the Tshikuwi dip tank; and soil at the Tshituni dip tank is gravelly, with traces of brown clay.

Sampling and testing

The points where soil samples were collected to investigate the horizontal distribution of arsenic were within a radius of 100 m from each tank. A normal dip site in communal areas covers an area of approximately 1 ha. The level of arsenic concentration was measured at distances of 5 m, 20 m and 100 m from the respective dip tanks. The 5-m collection site is the splash area and close to the poison hole where the solution is discarded when the tanks are cleaned (Figure 1). The 20-m distance covers a draining pen in which the cattle cluster whilst still wet with dip solution. The 100-m distance covers a radial area around the tanks

Table 1: Locations and features of 10 dip tank sites in the Vhembe district, Limpopo Province of South Africa

Sites	Latitude	Longitude	Established	Ecoregions and soil
Established before 1948				
Tshivhulani	22 55.35 S	30 30.12 E	Early 1920s	Ecoregion 2.01 (central highland): deep red clays predominate
Khubvi	22 49.52 S	30 34.03 E	1923	Ecoregion 2.01 (central highland): heavily weathered, compacted red clay
Rambuda	22 47.05 S	30 27.06 E	1940	Ecoregion 5.04 (northeastern area): red loam with high content of organic matter
Tshikuwi	22 53.83 S	29 58.91 E	1940	Ecoregion 5.03 (western area): heavily weathered, compacted red loam
Tshituni	22 56.82 S	30 02.57 E	1940	Ecoregion 5.03 (western area): gravelly with traces of brown clay
Established from 1948 onwards				
Sambandou	24 49.59 S	30 39.33 E	1948	Ecoregion 5.04 (northeastern area): sandy loam with very high content of organic matter
Tshifudi	22 48.24 S	30 43.27 E	1948	Ecoregion 2.01 (central highland): sandy loam with organic matter prevalent
Mukula	22 51.00 S	30 36.59 E	1948	Ecoregion 2.01 (central highland): weathered, compacted red clay
Thengwe	22 49.59 S	30 32.58 E	1950	Ecoregion 5.04 (northeastern area): sandy with little organic matter
Tshandama	22 30.07 S	30 45.05 E	1950	Ecoregion 5.04 (northeastern area): sandy with little organic matter

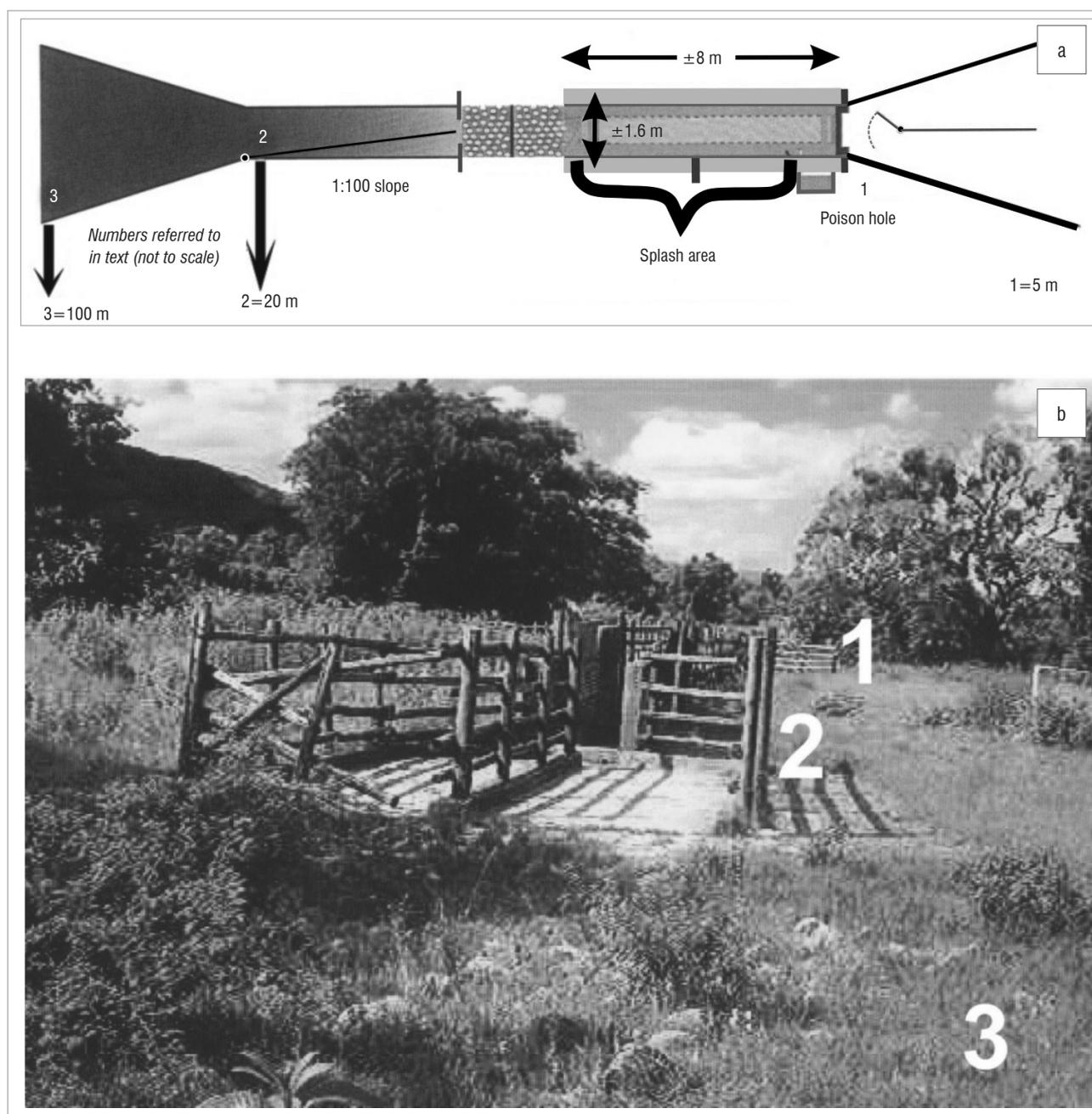


Figure 1: (a) Schematic of the aerial view of the dip tank layout and (b) a photo of a dip tank showing the sampling distances at points 1 (5 m), 2 (20 m) and 3 (100 m).

from where the cattle disperse. The 100-m distance was used as the control site.

Single, linear point soil samples following the contours of the terrain were taken at the sampled dip sites. The samples were taken at a depth of 300 mm, and placed in clean, labelled plastic bags.

The packaged soil samples were chemically analysed for arsenic by the accredited soil laboratory of the Agricultural Research Council. The analysis was performed using a semi-quantitative scan of an ammonium EDTA extract. An ammonium EDTA solution was added to soil samples, and the solution was filtered to isolate the chemicals.

Results and discussion

Figure 2 displays the average levels and spatial distribution of arsenic at the 10 dip sites.

Absolute concentration of arsenic residues

Surface soil around the 10 dip sites depicted enhanced arsenic values (Table 2), which ranged from 0.001 mg/kg to 46.76 mg/kg at a 5-m distance from the tanks (Figure 2). Sambandou had the highest mean concentration (18.24 mg/kg) and Tshandama the lowest (0.002 mg/kg). Five of the 10 dip sites (Khubvi, Rambuda, Sambandou, Tshifudi and Tshivhulani) displayed moderate to high concentrations (>3 mg/kg) of arsenic residues at the 5-m distance.

A first observation is that the arsenic concentrations measured in this study are definitively lower than those found in the 2007 study at the Luvuvhu River.²⁸ When excluding other variables, the difference in arsenic concentrations may be explained by the depth at which soil samples were extracted. While Moremedi and Okonkwo²⁸ took their samples at a maximum depth of 100 mm, samples for the purpose of our study were taken at a depth of 300 mm.

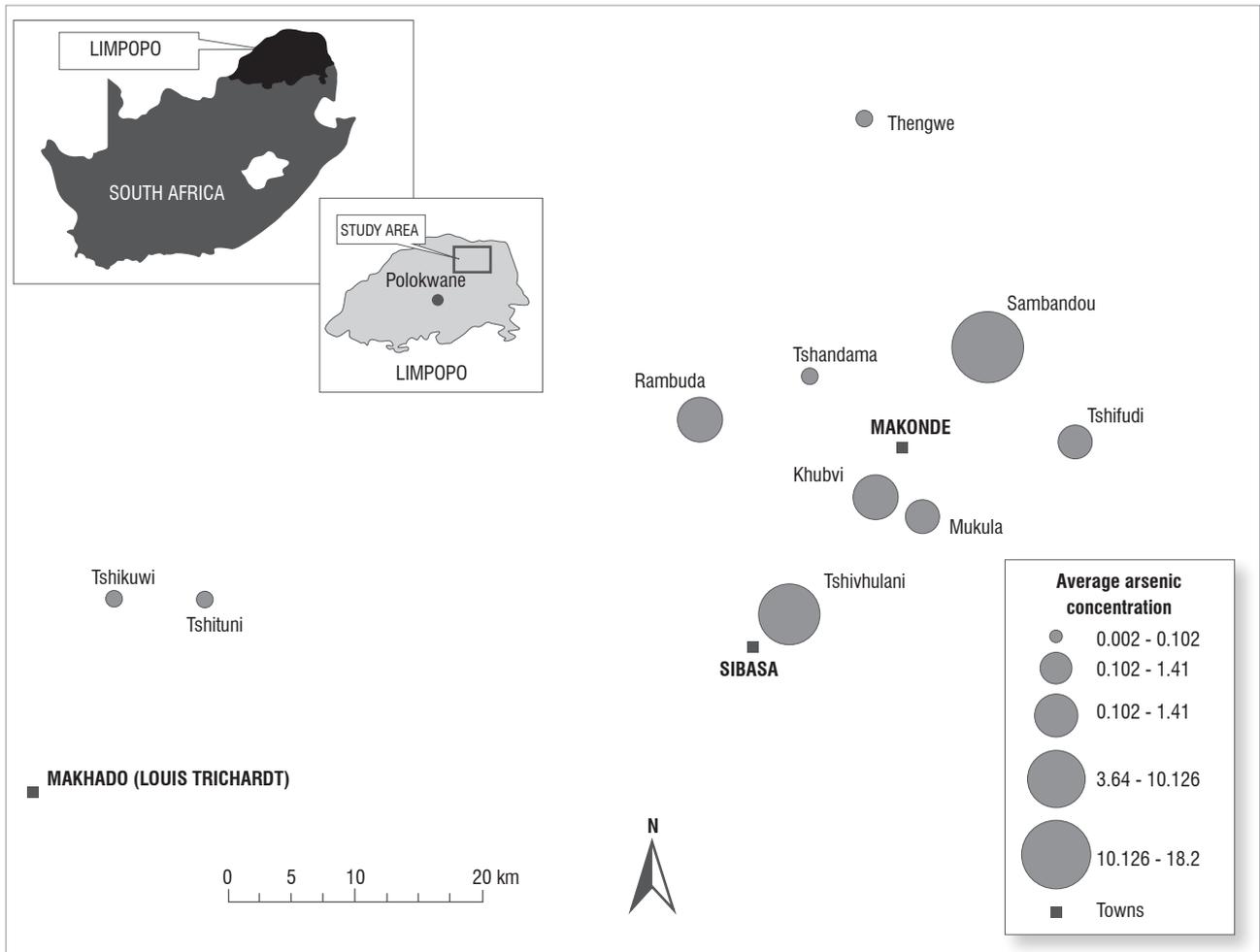


Figure 2: Average arsenic concentrations (mg/kg) in soil at 10 dip sites in the Vhembe district, Limpopo Province, South Africa.

Table 2: Concentration[†] of arsenic residues in mg/kg at the 10 dip tank sites

Dip site	5 m (A)	20 m (B)	100 m (control) (C)	Mean = A+B+C/3	Trends		
					B-A	C-B	C-A
Sambandou	46.76	6.88	1.09	18.24	-39.88	-5.79	-45.67
Tshivhulani	30.18	0.19	0.01	10.12	-29.99	-0.18	-30.17
Rambuda	3.53	3.63	3.70	3.62	0.10	0.07	0.17
Khubvi	3.65	3.69	3.60	3.65	0.04	-0.09	-0.03
Tshifudi	3.85	0.23	0.15	1.41	-3.62	-0.08	-3.7
Mukula	2.30	1.20	0.08	1.19	-1.1	-1.12	-2.22
Thengwe	0.14	0.07	0.09	0.10	-0.07	0.02	-0.05
Tshikuwi	0.08	0.12	0.02	0.07	0.04	-0.1	-0.06
Tshituni	0.02	0.06	0.01	0.03	0.04	-0.05	-0.01
Tshandama	0.002	0.003	0.002	0.002	0.001	-0.001	0.0
Mean	9.05	1.61	0.88		-7.44	-0.73	-8.17

[†]Values rounded to two decimal places.

Relative concentration of arsenic residues

The relative distribution of arsenic residues can be explained by a number of factors, namely duration of exposure, distance from dip tank and the properties of the receiving soil.

Duration of exposure

Duration of exposure only partly explains the differences in arsenic concentrations. If 1955 is taken as the year when the use of arsenic-based dipping compounds was discontinued, the average exposure of dip sites for which the five lowest concentrations were detected is 9.4 years ($\sigma=4.63$) and that of the tanks for which the five highest concentrations were found is 18.6 years ($\sigma=11.32$). At Sambandou, where the highest readings (46.76 mg/kg at 5 m and 6.88 mg/kg at 20 m) were recorded, the dip was in use for only 7 years before 1955, compared to approximately 32 years for the site with the second highest readings, Tshivhulani. The tanks constructed before 1948 ranked second, third and fourth for arsenic concentrations at the 5-m distance.

Distance

Arsenic contamination was generally higher closer to the tanks (Figure 3). However, the decline in contamination with distance varied. Concentrations at Sambandou (the site with the highest mean) declined by 39.88 mg/kg from the 5-m to the 20-m location, and by a further 5.79 mg/kg from the 20-m to the 100-m distance. The concentration at 5 m was about 43 times higher than that at 100 m. At Tshivhulani, the site with the second highest mean concentration, the measurement declined by 29.99 mg/kg from the 5-m to the 20-m distances and by a further 0.18 mg/kg from the 20-m to the 100-m distances. At this site, the concentration at 5 m was more than 3000 times higher than the concentration at 100 m. A trend of declining arsenic concentrations with distance was also found at two other sites – Tshifudi and Mukula. The high levels of contamination closer to the tanks could be attributed to

two factors: firstly, the practice for many decades of disposing of spent dip sludge by discarding it onto the ground or into nearby ‘poison holes’ when the tanks were cleaned, and, secondly, splashing of dip solution over the sides of the dip tanks every time cattle entered the tanks.

Measurements at the other six sites revealed different patterns. At Khubvi (ranking third in mean concentrations), the concentration first increased from the 5-m to the 20-m point before it declined at the 100-m point, with an overall decline of 0.03 mg/kg from 5 m to 100 m. Concentrations measured at Rambuda (with the fourth highest mean concentration) showed a general increase of arsenic levels with distance, although the overall increase was small (0.17 mg/kg from 5 m to 100 m). The difference from the overall pattern of declining arsenic concentrations with distance at Khubvi and Rambuda tanks could be attributed to human activity: the area around Khubvi tank has subsequently been turned into a maize field, whilst an area adjacent to the Rambuda tank is used for the manufacture of mud bricks. These activities might have shifted the soil downslope, and, consequently, assisted in the migration of arsenic. In addition, the poison hole at Rambuda tank is situated at its lower side, approximately 20 m away from the tank, and is joined to the tank by a narrow furrow. As a result, dip effluent was prevented from soaking into the ground before it reached the poison hole.

Soil properties

It is accepted worldwide that there is a positive correlation between relatively higher arsenic concentrations and clay, silt and organic matter and, specifically, iron and aluminium oxides in soil.^{2,5,6,33} A high concentration of arsenic is mainly found in the top layers in areas with clayey soil. By contrast, arsenic is easily leached or washed into deeper layers in areas where large-grained sandy soil occurs, because of the lower absorption capacity of this type of soil. Flooding and weathering also appear to enhance the horizontal distribution of arsenic in soil, with

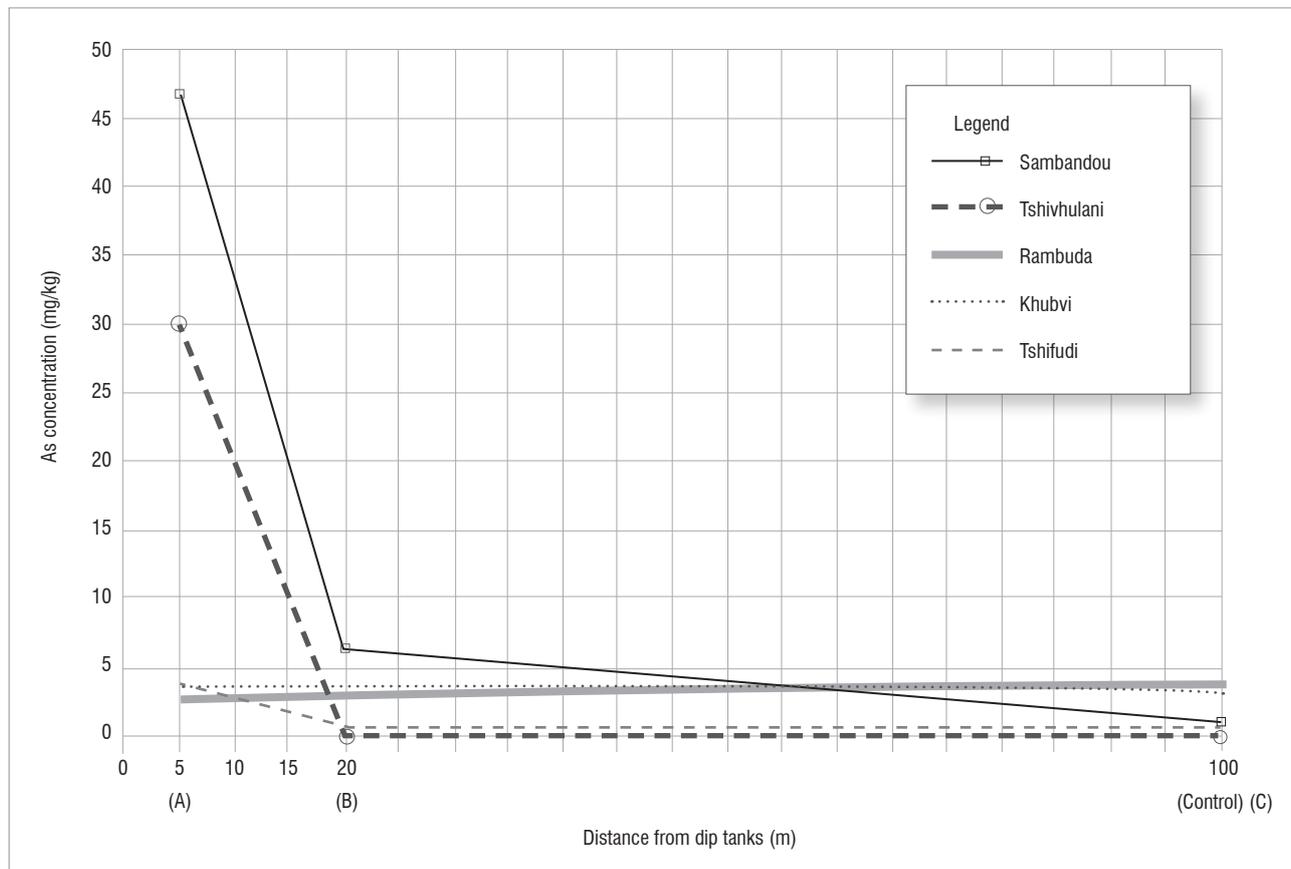


Figure 3: Arsenic concentration in soil at 5-m, 20-m and 100-m distances from dip tanks at five sites in the Vhembe district, Limpopo Province, South Africa.

both of these processes contributing to the deeper penetration of arsenic into the soil profile.³⁴

The findings of our study generally confirm previous observations, that is, either clay or a high content of organic matter were prevalent at the five sites with the highest arsenic concentrations (Table 3). The loamy and clay rich soil at Khubvi, Rambuda, Sambandou, Tshifudi and Tshivhulani had higher arsenic concentration levels than Tshandama, Thengwe, Tshikuwi and Tshituni, where sandy and rocky soil is more dominant.

Table 3: Arsenic concentrations at 5 m from the dip tank and surrounding soil characteristics

Arsenic (mg/kg) at 5 m	Site	Soil characteristics
46.76	Sambandou	Sandy loam with very high content of organic matter
30.18	Tshivhulani	Deep red clay
03.85	Tshifudi	Sandy loam with prevalent organic matter
03.65	Khubvi	Heavily weathered, compacted red clay
03.53	Rambuda	Red loam with high content of organic matter

Summary, discussion and conclusion

We investigated the arsenic contamination of soil at cattle dip tanks in the Vhembe district of Limpopo where arsenic-based dipping compounds were used from the 1910s to the mid-1950s. The main findings of the study are:

- High concentrations of arsenic residues (>3 mg/kg) were found at a depth of 300 mm at a number of dip tank sites, namely Khubvi, Rambuda, Sambandou, Tshifudi and Tshivhulani. It is assumed that arsenic concentrations could be much higher at shallower depths.
- Differences in the concentrations at the 10 dip sites are ascribed to (1) the period of exposure to arsenic-based dipping compounds (with dip tanks constructed before 1948 generally having a higher level of contamination than dip tanks constructed after this date), and (2) soil properties (with clay soils and high levels of organic matter correlating with higher arsenic concentrations).
- A decline in arsenic concentration with distance from the dip tanks was evident, indicating that soil contamination is spatially localised.
- The contribution of inorganic arsenic-based dipping compounds to arsenic concentrations in the soil is evident, with mean values of 9.05 mg/kg at 5-m distances compared to 0.88 mg/kg at 100-m distances from the 10 dip tanks.

The detected levels of arsenic contamination are concerning. The first and immediate danger is direct human contact with contaminated sites. Of particular concern are encroaching villages, the health of dip operators and water resources. None of the sites referred to in this study were fenced. In the cases of Sambandou (46.76 mg/kg at 5 m) and Rambuda (3.53 mg/kg at 5 m), the nearest houses were 70 m and 30 m from the dip tanks, respectively. Children were observed playing in the vicinity of the dip tanks and they often assisted with dipping operations without skin protection. Oral accounts by informants confirmed that throughout various decades, and even until recently, dip assistants had not received proper training on the dangers of the chemicals applied during dipping and seldom wore protective clothing. Water courses, wells, springs and boreholes could also be threatened because of the soluble nature of arsenic-based dipping compounds. In addition, there is a danger of spillage and spread of residues through flooding. The Tshivhulani dip tank is, for example, only 20 m from the nearest water course. Moreover, a government report indicated that already in 1951 several dip tanks, including those in Mukula, Rambuda, Tshifudi and Tshivhulani, needed

urgent repairs.³⁵ Our inspection revealed that dip tanks in the district are generally in a poor structural condition with most showing deep cracks. Crops are being cultivated closer and closer to dip tank sites, and, in the case of Rambuda, the tank is located within the boundary of a crop field.

However, the possible transfer of arsenic residues into the food chain should be treated with caution. It depends on a variety of factors including the total arsenic in soil (bioavailability), bioaccessibility of the substance, transfer of soil arsenic to the edible parts of plants (bioaccumulation), and the human intake of arsenic.^{5,36} Although clay and organic matter have a higher adsorbing capacity for arsenic than sand, these materials are weakly correlated with bioaccessibility as a result of the bonding strength of soil particle bound arsenic.¹³ Sheppard³³ concluded that inorganic arsenic was five times more toxic to plants in sand than in clay, because sandy soil generally contains lower amounts of iron and aluminium oxides than clayey soils. Thus, although certain crops, such as rice, display higher bioaccumulation of arsenic than other crops, care should be taken in correlating arsenic concentrations in soil with a perceived presence in food sources.^{2,5}

In conclusion, although the eradication of ECF was achieved and foot and mouth disease prevention was facilitated, the ecological legacy of arsenic-based dipping compounds still lingers in the communal areas of South Africa. A widespread, detailed investigation in communal areas, taking into account the factors identified in this study, should be undertaken to provide more detail about the health threats of arsenic contamination at dip sites in the country.

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Authors' contributions

M.R.R. was the principle researcher, conducted the fieldwork and supervised the laboratory testing. A.C.H. extended the literature survey, helped with the interpretation of the data and prepared the final version of the manuscript.

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