

## Research Article

# Soil Heavy Metal Distribution with Depth around a Closed Landfill and Their Uptake by *Datura stramonium*

Peace Makuleke and Veronica M. Ngole-Jeme 

Department of Environmental Science, School of Ecological and Human Sustainability,  
College of Agriculture and Environmental Sciences, UNISA, Florida 1710, Roodepoort, Gauteng, South Africa

Correspondence should be addressed to Veronica M. Ngole-Jeme; [vm.ngole@gmail.com](mailto:vm.ngole@gmail.com)

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Landfills are major sources of environmental pollution. This study evaluated heavy metal concentrations in soils and plants around the closed Lumberstewart landfill in Bulawayo, Zimbabwe, to determine the pollution potential of a closed landfill and the risks they present to plants growing in this environment and surrounding communities. Soil samples were collected at depths of 0–30 cm, 30–60 cm, and 60–90 cm around the landfill and at a control site and characterized for various properties and concentrations of Cd, Cu, Cr, Fe, Ni, and Zn. Samples of *Datura stramonium*, collected from the same sites where soil samples were collected, were also analyzed for the same heavy metals. The soils were sandy, mostly acidic ( $5.01 < \text{pH} < 7.65$ ) with low organic matter content ( $< 2\%$ ) and cation exchange capacity ( $< 15 \text{ meq}/100 \text{ g}$ ). These properties varied with depth around the landfill. Heavy metals concentrations in the soils and *Datura stramonium* followed the order  $\text{Fe} > \text{Zn} > \text{Cu} > \text{Cr} > \text{Ni} > \text{Cd}$  with samples from around the landfill having higher concentrations than samples from the control site. Soil heavy metal enrichment was highest at a depth of 30–60 cm. Pollution load index (PLI) values indicated that all sites around the landfill were polluted ( $\text{PLI} > 1$ ). Heavy metal transfer coefficient in *Datura stramonium* ranged between 0.0 and 209 with  $< 60\%$  of the variation observed in heavy metal transfer coefficient in *Datura stramonium* explained by the extent of heavy metal enrichment in the soils. More than 20 years after closure of the landfill, there are indications that leachate migration may still be going on around the landfill. Monitoring of environments around closed landfills needs to be ongoing to mitigate negative impacts on humans and the environment.

## 1. Introduction

Municipalities, commercial centers, and industries worldwide generate tons of waste annually which end up in landfills. Landfills remain the best option for solid waste disposal because they can accommodate huge volumes of waste and are cheaper to run compared to other solid waste disposal options such as incineration. Despite these advantages, landfills remain major point sources of pollutants such as heavy metals and organic compounds to the environment. Landfill leachate produced from a combination of decomposing waste and liquids percolating through waste in landfills is the main vector of these contaminants from the landfill to surrounding environments. According to Aziz et al. [1], Kamaruddin et al. [2], Kjeldsen et al. [3], and Ngole

and Ekosse [4], landfill leachate usually contains dissolved organic matter, inorganic macrocomponents, heavy metals, and xenobiotic compounds derived from decomposing waste. Among these leachate constituents, heavy metals and xenobiotic compounds have received global attention because they cause several negative health and environmental impacts.

Heavy metals describe a group of elements that occur naturally and have atomic number, density, and molecular weight greater than 20,  $6 \text{ g}/\text{cm}^3$ , and 53, respectively [5, 6]. They are used for a variety of applications and products and are therefore contained in waste entering most landfills. The occurrence of heavy metals such as Cd, Cr, Cu, Pb, Ni, Fe, and Zn in landfill leachate and soils around landfill environments is widely reported [4, 7, 8] and is associated with

the migration of landfill leachate. Leachate production in, and migration from landfills to surrounding environments continue even after the landfill is closed especially if the landfill was not properly capped before closure. Studies by Kim and Lee [9] and Robinson [10] have highlighted the risks associated with leachate in landfills that have been closed for over 6 years. The extent of leachate and pollutant migration from both closed and active landfills is a function of the properties of soils in the surrounding environment. Soil texture determines the rate at which landfill leachate infiltrates into the surrounding environments, whereas soil organic matter content, cation exchange capacity, mineralogical composition, and pH dictate the rate of pollutant dispersion [11–13].

Closed landfill sites remain as available spaces for various developmental and recreation projects. Golf courses, recreation parks, playgrounds, animal refuge, tennis court, and industrial site are some of the land use types found on closed and capped landfills [14, 15]. They have also been used as grazing fields for livestock and as areas where solar panels are set [16]. Soils around landfills are usually fertile and are sometimes used for growing food crops [15]. Where not in use, closed landfills are colonized by weeds, some of which have medicinal properties. Monitoring and maintaining a closed landfill for a period of at least 30 years has been recommended to decrease the negative impacts associated with migrating leachate from such landfills [15]. However, the level of adherence to this environmental safety guideline reduces as the period post closure of the landfill increases. There is therefore a possibility that communities around closed landfills may be making use of these environments without any knowledge of the soil quality around the landfill or the suitability of the soil for various purposes.

Studies by Othman et al. [17] have reported continuous migration of leachate from closed landfills. However, the rate of migration decreases by 75% one year after closure and up to 90% after 4 years [18]. Over time, the organic chemical substances contained in the leachate are degraded or leached out of the landfill with water, but heavy metals remain because of their nonbiodegradable nature. Soils around closed landfills may contain high concentrations of heavy metals and other contaminants derived from the leachate with dire consequences on the human and physical environment. A decrease in the population and distribution of soil microbes and microbial enzymatic activities has been associated with elevated soil heavy metal contaminations [12, 19, 20]. Heavy metals affect plant growth including fluorescence, stomatal resistance, oxidative stress, reproductive processes, seed morphology and physiology, and seed germination [21]. Research indicates that vegetables and other food crops grown around closed landfills accumulate high levels of heavy metals and other contaminants from soils contaminated by landfill leachate [22, 23]. Ingestion of vegetables that has taken up heavy metals present significant health threats to the consumer. Human exposure to heavy metals is incriminated in several medical conditions including poisoning of the skeletal, nervous, respiratory, excretory, and digestive systems, skin damage, nasal and lung irritation, ulceration of the stomach and small

intestines, decreased sperm count, and kidney disease [24–26]. According to Ngole-Jeme and Fantke [27], heavy metals are potential carcinogens.

Zimbabwe, like many developed and developing countries, has a challenge of managing waste quantities, waste disposal, and waste impacts on the environment. Lumberstewart landfill in Bulawayo, Zimbabwe, was in use between 1978 and 1990 when there were no regulations governing the design and use of landfills in Zimbabwe. The landfill is currently colonized by weeds among which are Jimson weed (*Datura stramonium*) and pigweed (*Amaranthus* family), which are used for various medicinal purposes in Zimbabwe [28] and India [29] to alleviate headaches and symptoms of asthma, bronchitis, and other ailments. High concentrations of heavy metals in these weeds could impact its beneficial use as it could become a source of human exposure to heavy metals creating more health challenges rather than solving them. No research has been performed to establish whether the concentrations of potentially toxic heavy metals Cd, Cr, Cu, Pb, Ni, Fe, and Zn in soils around Lumberstewart landfill are elevated more than 20 years after it was closed or whether the plants that are growing around the landfill are taking up these heavy metals. The aim of this study was to assess heavy metal concentrations in soils around the closed landfill in an endeavor to understand the extent of heavy metal contamination in the area years after the closure of the landfill. It also aimed to determine whether *Datura stramonium* growing around the closed landfill is taking up heavy metals from the soil and the potential health threat that this may pose to communities using the weed for medicinal purposes.

## 2. Site Description

The closed Lumberstewart landfill is located on the highveld in the western region of Bulawayo City in Zimbabwe at latitude 20°13' 55" S and longitude 28°56' 39" E (Figure 1). The landfill covers an area of approximately 300 m × 200 m and has an altitude of 1358 m above sea level. Climate in the area is subtropical due to its high altitude. Mean annual temperature of the area is 19.6°C, and October is usually the hottest month with temperatures of over 40°C, whereas the coldest month is July with minimum temperatures averaging 7°C [30, 31]. Bulawayo city receives low and erratic precipitation with an average annual rainfall of about 600 mm [30]. The dominant rock type around the landfill is granitic in nature with quartz, feldspar, and mica as the dominant primary minerals [32]. The soils are acidic and sandy, with low organic matter content [30]. The landfill was not lined but had compacted clay at the base to minimize seepage of leachate into the subsurface environment [32]. There was no leachate or methane collection system to collect leachate generated from the landfill or methane gas emitted through anaerobic waste decomposition in the landfill, respectively. Upon its closure, the landfill was capped with compacted sand and coal ash. Lumberstewart landfill used to be the main disposal site for the entire city before its complete closure in 1990.

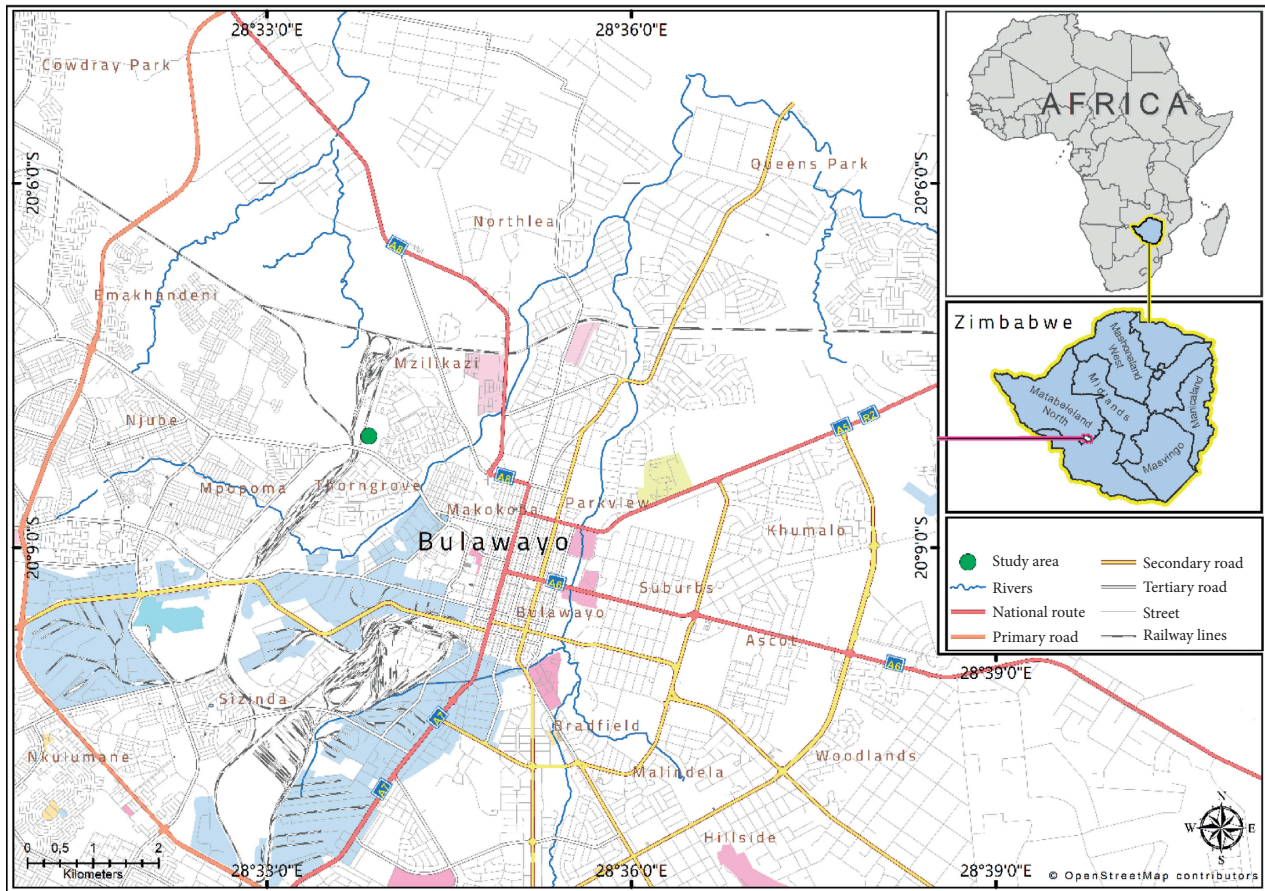


FIGURE 1: Location map of Lumberstewart landfill.

### 3. Research Methods

**3.1. Collection of Soil and Plant Samples.** A systematic sampling approach was used to collect soil samples around the landfill. Three soil sampling sites were located along each of the northern and southern landfill boundaries and four each along the western and eastern boundaries giving a total of 12 sampling sites around the landfill. At each boarder of the landfill, the sampling points were evenly spaced. About 500 gm of soil sample was collected at depths of 0–30 cm, 30–60 cm, and 60–90 cm at each of the sampling points and a control site located 2 km from the landfill using a steel auger and hand trowel. A total of 39 soil samples were therefore collected from all 13 sites. The soil samples were hot air-dried at 35°C after which they were disaggregated with a wooden pestle and porcelain mortar, passed through a 2 mm sieve, and analyzed for the different parameters. Samples of *Datura stramonium* were collected from the same sites where soil samples were collected. The plant samples were washed with distilled water to remove soil particles after which they were dried in a well-ventilated oven at 60°C for 48 hours. The dried samples were ground, and the heavy metals content in them was determined [33].

**3.2. Samples Analyses.** Physicochemical properties analyzed in the soil samples included pH, electrical conductivity (EC), texture, organic matter (OM), and cation exchange

capacity (CEC). The texture of the soil was determined using the Bouyoucos hydrometer method [33]. Soil pH was measured in a 1:2.5 (weight: volume) soil:water suspension [32] using a microprocessor HANNA Instrument pH meter model HI2002. A calibrated HANNA Instrument EC meter model HI2003 was inserted in the same suspension used for pH measurement after letting the suspension to stand for about an hour to determine soil EC [34]. Both pH and EC meters used were of South African make. To determine the CEC of soil samples, the ammonium acetate method as described by Ngole and Ekosse [4] was used. The OM content in the soil was determined using the loss-on-ignition (LOI) method as per procedure outlined by Luke et al. [35]. In this procedure, the sample was first dried at 105°C to eliminate water molecules after which a known weight was fired in a muffle furnace at 550 for 2 h. The percentage decrease in weight of the samples after heating in the muffle furnace was assumed to be the amount of organic matter contained in the samples.

To determine the concentrations of Cd, Cr, Cu Fe, Ni, and Zn in the soil, soil samples were digested according to the EPA Method 3050B [36]. 1 g of each soil sample was weighed into a porcelain crucible and 10 ml of a 1:1 mixture of HNO<sub>3</sub> and HF was added. The crucible was then allowed to dry in a waterbath after which 20 ml of 2 M HNO<sub>3</sub> was used to dissolve the residue. Particles in the resulting solution were removed by filtration using an acid-washed

Whatman No. 41 filter paper [37]. The solution was then transferred to a 100 ml volumetric flask and made up to volume using distilled deionized water. Heavy metal concentration in the resulting solution was determined using the German Analytik Jena ContrAA 300 flame atomic absorption spectrometry [37, 38].

The concentrations of Cd, Cr, Cu, Fe, Ni, and Zn in the plant samples were determined by digesting the samples with nitric acid (HNO<sub>3</sub>) and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) [33]. In this procedure, 5 g of each ground plant sample was weighed into a conical flask after which 5 ml of 65% HNO<sub>3</sub> and 5 ml of 30% H<sub>2</sub>O<sub>2</sub> were added. The mixture was heated on a hot plate at 140°C until the contents had reduced to 5 ml [33, 39]. The contents were filtered through an acid-washed filter paper, transferred to a 50 ml volumetric flask, and made up to volume with deionized water. The ContrAA 300 flame atomic absorption spectrometer was then used to determine the concentrations of Cd, Cr, Cu, Fe, Ni, and Zn in the plant digest.

**3.3. Quality Control and Quality Assurance Measures.** All samples were collected and analyzed in duplicate. Values presented for the different parameters are therefore means of two samples from each of the sampling site. Glasswares were soaked in 1% nitric acid for 24 hours prior to their use. The reagents used were all Analar grade reagents. In the determination of heavy metals, high purity metal standards approved and endorsed by National Institute of Standards and Technology (Fluka \* Analytical Sigma-Aldrich, Germany) were used for calibration, and calibration checks were performed after every five samples [40]. Heavy metal recovery was determined using standards with known concentration of the different heavy metals. Equipment conditions were adjusted until a heavy metal recovery percentage of between 87% and 108% was achieved using the known standards.

**3.4. Data Analyses.** Descriptive statistics including mean and standard deviation of the different soil properties and heavy metal concentrations in the samples were determined using SPSS version 25.0. ANOVA with Tukey's test of significance was used to separate means of heavy metal concentrations and soil properties between sites and between depths at the same site. All statistical analyses were carried out at a 95% confidence interval. Significant differences were therefore reported at a probability (*p*) value of 0.05. To determine the extent of heavy metal contamination in soils at the landfill, heavy metal contamination factor (CF) for each of the heavy metals analyzed was calculated according to equation (1) as recommended by Hakanson [41] and Ngole-Jeme [42]:

$$CF = \frac{C_{m \text{ sample}}}{C_{m \text{ background}}}, \quad (1)$$

where  $C_{m \text{ sample}}$  is the concentration of a metal in soils from the landfill, and  $C_{m \text{ background}}$  is the concentration of same heavy

metal in background samples (which in this case was the sample from the control site).

According to Sutherland et al. [43], values of heavy metal  $CF < 1$  refer to low contamination,  $1 \leq CF < 3$  implies moderate contamination,  $3 \leq CF \leq 6$  indicates considerable contamination, and  $CF > 6$  indicates very high contamination. The pollution load index (PLI) which indicates the extent of heavy metals contamination in soils was also calculated to determine which of the sites was the most contaminated when all heavy metals analyzed are taken into consideration [44]. Pollution load index values  $< 1$  indicate no pollution, whereas values  $> 1$  indicate pollution [45]. The PLI for each site was calculated according to Seshan et al. [45] and Tomlinson et al. [46] as shown in the following equation:

$$PLI = n\sqrt{CF_{Cd} \times CF_{Cr} \times CF_{Cu} \times CF_{Fe} \times CF_{Ni} \times CF_{Zn}}, \quad (2)$$

where CF is the contamination factor, and *n* is the number of elements which in this study were 6.

The mean values of the concentrations of the different heavy metals in the plant samples from the landfill and those from the control site were compared. Heavy metal transfer factor between the soils and *Datura stramonium* growing on the landfill and control sites was determined as indicated in the following equation [47].

$$T.F = \frac{C_{plants}}{C_{soils}}, \quad (3)$$

where T.F is the transfer factor,  $C_{plants}$  is the heavy metal concentration in plants, and  $C_{soils}$  is the heavy metal concentration in soil.

Transfer factors for the different heavy metals in *Datura stramonium* were correlated with the heavy metals contamination factor of the soils at a depth of 0–30 cm to identify any relationship that existed between the extent of heavy metal enrichment of the soil and the uptake of heavy metals by the plants.

## 4. Results and Discussion

**4.1. Properties of Soils at the Landfill.** Soils from the landfill had sand, clay, and silt contents ranging between 61.6% and 76.5%, 11.5% and 23.2%, and 9.30% and 17.1%, respectively. The control site had a higher percentage of sand (81.4%) compared to sites around the landfill, but its clay content was less, averaging 5%. No variation in the content of sand, silt, and clay was observed with depth around the landfill, and so, the texture of the soils was similar at all depths, falling within the sandy loam and sandy clay loam textural classes (Figures 2(a)–2(c)). Soils belonging to these textural classes are characterized by high porosity and permeability, which may facilitate leachate migration into surrounding environments.

The soils may also have low sorption capacity because of the low clay content. According to Madzhieva et al. [48], the sorption of heavy metals is lower in coarse-textured soils than in fine-textured soils because of the

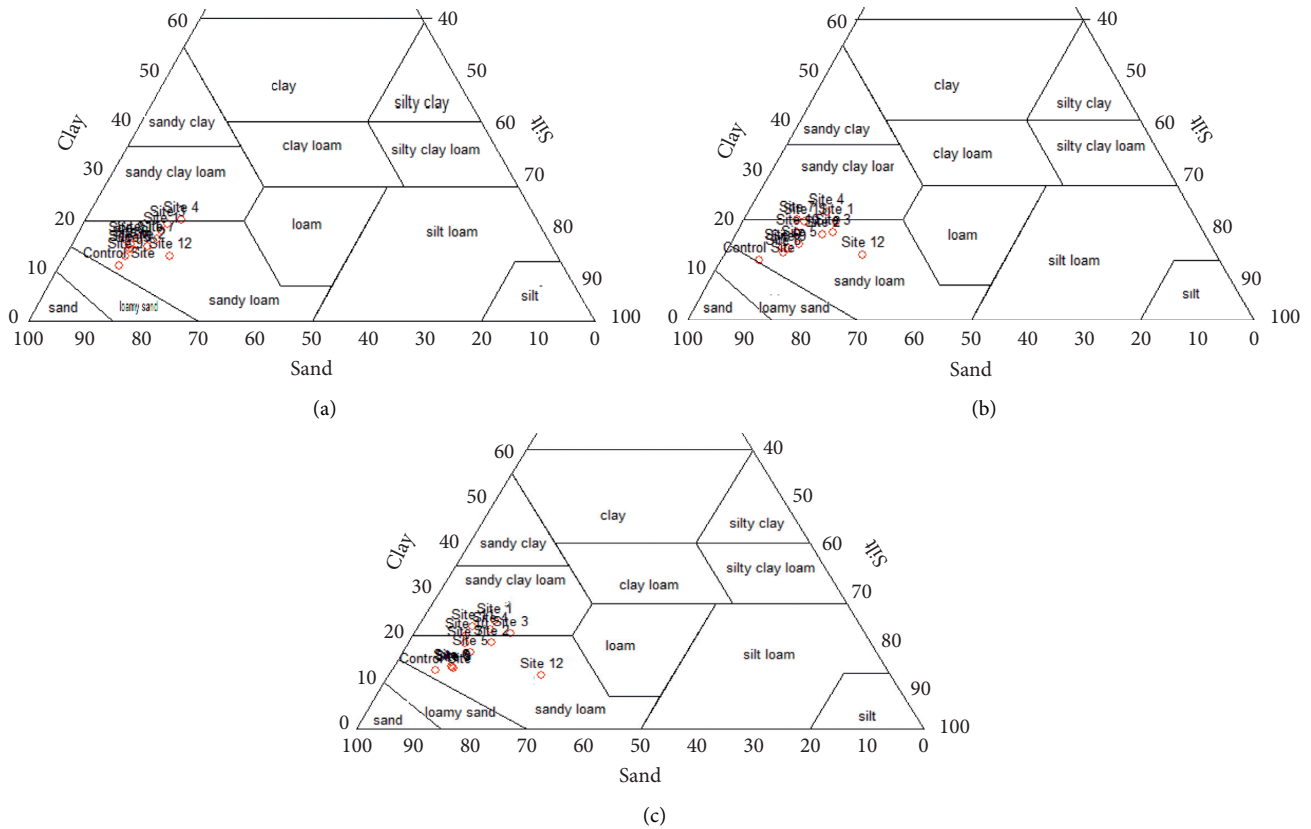


FIGURE 2: Textural triangles showing the texture of the soils at depths of 0–30 cm (a), 30–60 cm, (b) and 60–90 cm (c) at the landfill.

lower surface charge density and specific surface area of coarse-textured soils compared to finer soils. The texture of the soils around the landfill could therefore contribute towards the movement of leachate and dispersion of heavy metals from the closed landfill into surrounding environments.

The pH of soils around the landfill ranged from 5.01 to 7.65 with many of the samples having pH values below 7.0 (Figure 3). These values were slightly lower than what was obtained at the control site with pH values between 7 and 7.6. Differences in pH between the soils from the landfill and those from the control site were, however, insignificant ( $p > 0.05$ ). The slightly acidic nature of the soils in this study is similar to those reported by Salami et al. [49] but contrary to what was reported by Beyene and Banerjee [50] in other landfills. Leachate in old landfills is generally believed to be alkaline in nature [51] and are expected to increase the pH of environments into which they migrate. However, Seo et al. [52] have attributed acidic pH in old landfills to the presence of bicarbonate salts in the leachate. The low pH values observed in some soils around the landfill are therefore not unexpected. There were variations in soil pH with depth around the landfill with pH values of soils at a depth of 30–60 cm being significantly higher than what was obtained in soils at a depth of 0–15 cm ( $p = 0.02$ ) and 60–90 cm ( $p = 0.01$ ) (Figure 3). Metal mobility in soils generally increases with decrease in soil pH, whereas nutrient availability is high when soil pH is below 7 [53, 54]. The slightly

acidic nature of the soils around the landfill may have limited impact on metal mobility in soils around the landfill environment.

Electrical conductivity values of the soils ranged from 0.43 to 1.67 dS/m and increased with depth around the landfill (Figure 3). However, the differences in EC between depths were not significant ( $p > 0.05$ ). Some sites around the landfill had higher EC values than the control site which had values of 0.30 dS/m, 0.26 dS/m, and 0.15 dS/m at depths of 0–30 cm, 30–60 cm, and 60–90 cm, respectively. The EC values obtained in this study indicate that all sites had nonsaline conditions as the EC of soils at all sites were  $< 2$  dS/m [55]. Soil organic matter content around the landfill varied from one depth to the other (Figure 3). Similar to EC, some sites around the landfill also had significantly higher OM content than what was obtained in soils from the control site ( $p < 0.05$ ). Differences in OM content in soils at a depth of 0–30 cm and those at a depth of 30–60 cm were significant ( $p < 0.05$ ), whereas there were no differences in OM content between soils at depths of 30–60 cm and 60–90 cm ( $p > 0.05$ ). High OM content in surface soils around the landfill is not likely to be caused by landfill leachate migration from the landfill because the organic compounds in the leachate would have degraded over time. It could have accumulated because of residues from dead animals, plants, and plant roots since the site has been colonized by weeds. According to Tripathi and Misra [56], high OM at the surface is mainly because of the presence of organic residues

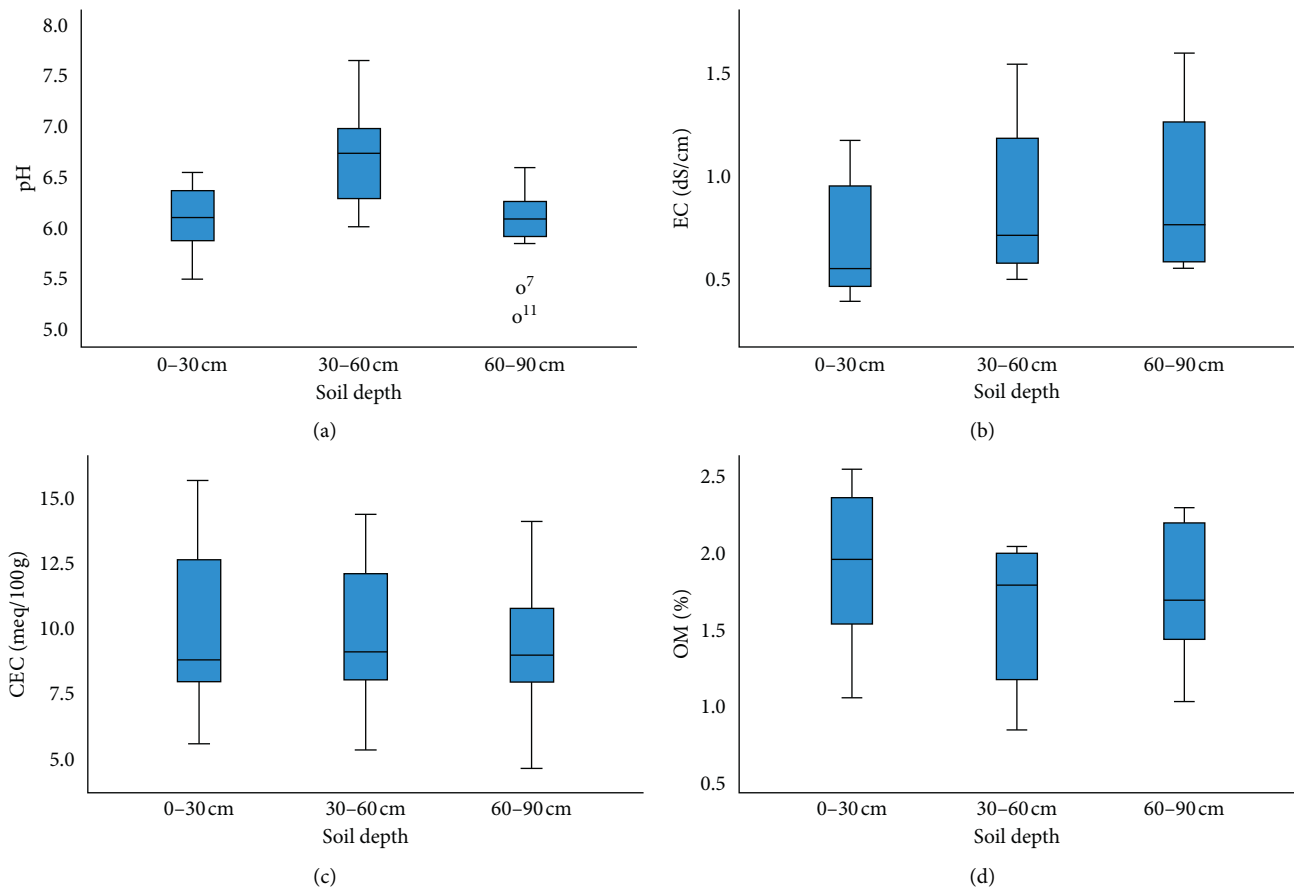


FIGURE 3: Physicochemical properties of soils around the landfill.

from agricultural activities and some recreational activities which add more OM to the soils.

The CEC of all soil samples collected from around the landfill and the control site were below  $20 \text{ cmol kg}^{-1}$ . For most of the sites, the values for CEC decreased with depth (Figure 3). According to Aydinalp and Marinova [57] and Khan and Kar [58], soil CEC increases with OM and clay content in soils. Though the soils had some amount of clay, the CEC values fall within the range of values reported for 2:1 clay minerals. The clay fraction of the soils is most likely dominated by these minerals which are characterized by low CEC. Organic matter content in the soils was also relatively low. Soil organic matter has pH-dependent charges which tend to be high when the soil is alkaline and low when soil pH is low due to protonation and deprotonation of functional groups contained in organic matter. Deprotonation caused by the acidic nature of the soils would have reduced the pH- negative charge density of the soil, reducing the fraction of CEC contributed by soil OM. The low CEC observed in the soils despite the high clay content could therefore have been caused by the mineralogical composition of the soils and their pH. The role of low soil CEC on high soil heavy metal mobility and their uptake by plants has been highlighted in several studies [59, 60]. Heavy metal migration and leaching of nutrients around the landfill are mostly like to be high due to the low CEC. This is compounded by the prevailing pH conditions which favor plant nutrient availability in the soils.

**4.2. Concentration of Heavy Metals in Soils around the Landfill.** The concentrations of Cd, Cr, Cu, Fe, Ni, and Zn in soils around the landfill varied with Fe having the highest concentration and Cd the lowest (Table 1). The concentration of Cd varied with depth (Figure 4), but the means at the different depths were similar as shown in Figure 4. The low concentration of Cd at all sites around the landfill including the control site may indicate naturally low concentrations of this heavy metal in the soils. Chromium concentration in soils from all landfill sites were significantly higher than what was obtained at the control site ( $p < 0.05$ ) (Table 1). High levels of Cr at the landfill compared to the control site have also been reported in other landfill site studies [8, 61] and have been attributed to leachate migration from the landfill. Though mean Cr concentrations were lower at the surface compared to the subsurface environment (Figure 4), the differences were insignificant ( $p = 0.79$ ).

The amount of Cu in soils at the landfill site was almost five times the concentration in soils from the control site (Table 1). Agyarko et al. [62] and Pasquini and Alexander [63] all found high concentrations of Cu in soils around landfills. Copper is used in the manufacture of several items including wires, vehicle parts, pipes, and alloys that find their way into the landfill. Leachate from most landfills is therefore expected to contain high concentrations of Cu derived from decomposition of these materials in the

TABLE 1: Mean concentrations of heavy metals in soils around the landfill at a depth of 0–90 cm.

Location	Site	Concentrations (mg/kg)					
		Cd	Cr	Cu	Fe	Ni	Zn
Eastern boundary	Site 1	0.40 ± 0.03	29.6 ± 3.38	127 ± 10.60	44716 ± 889	28.0 ± 1.97	288 ± 8.60
	Site 2	0.40 ± 0.00	25.6 ± 3.19	81.0 ± 12.60	41294 ± 744	35.0 ± 3.25	371 ± 8.30
	Site 3	0.37 ± 0.06	24.0 ± 2.70	78.2 ± 13.80	42309 ± 612	34.3 ± 3.34	342 ± 9.10
Northern boundary	Site 4	0.27 ± 0.07	23.8 ± 2.75	81.8 ± 14.40	39392 ± 857	31.8 ± 6.78	360 ± 5.20
	Site 5	0.11 ± 0.00	40.6 ± 4.40	93.6 ± 15.40	32622 ± 428	25.6 ± 3.37	267 ± 3.40
	Site 6	0.37 ± 0.09	41.9 ± 1.79	130 ± 9.70	45691 ± 802	28.9 ± 1.64	304 ± 7.50
Southern boundary	Site 7	0.51 ± 0.16	41.5 ± 3.53	129 ± 10.10	44694 ± 906	28.5 ± 1.80	297 ± 7.80
	Site 8	0.11 ± 0.00	39.6 ± 2.39	86.4 ± 8.80	33645 ± 855	22.0 ± 2.57	253 ± 4.70
	Site 9	0.11 ± 0.00	37.2 ± 1.16	79.0 ± 6.30	39796 ± 784	30.9 ± 2.68	349 ± 8.30
Western boundary	Site 10	0.11 ± 0.00	35.5 ± 1.55	63.9 ± 7.27	34849 ± 820	29.6 ± 3.48	333 ± 8.80
	Site 11	0.11 ± 0.00	35.3 ± 1.69	57.7 ± 8.08	34924 ± 924	29.4 ± 3.40	228 ± 8.10
	Site 12	0.11 ± 0.00	34.9 ± 1.52	69.9 ± 5.92	33238 ± 870	21.1 ± 2.88	273 ± 5.40
Control site		0.11 ± 0.00	11.7 ± 0.33	22.4 ± 1.81	12080 ± 439	19.7 ± 0.33	329 ± 8.14
*NEMA (1988)		7.5	6.5	16	NA	91	240
WHO		0.01	0.05	1.50	20.0	6.5	15.0
SABS, 1999		2	80	6.6	N/A	50	46.5

\*NEMA, National Environmental Management act (1998). \*\*SABS, South African Bureau of Standards 1999.

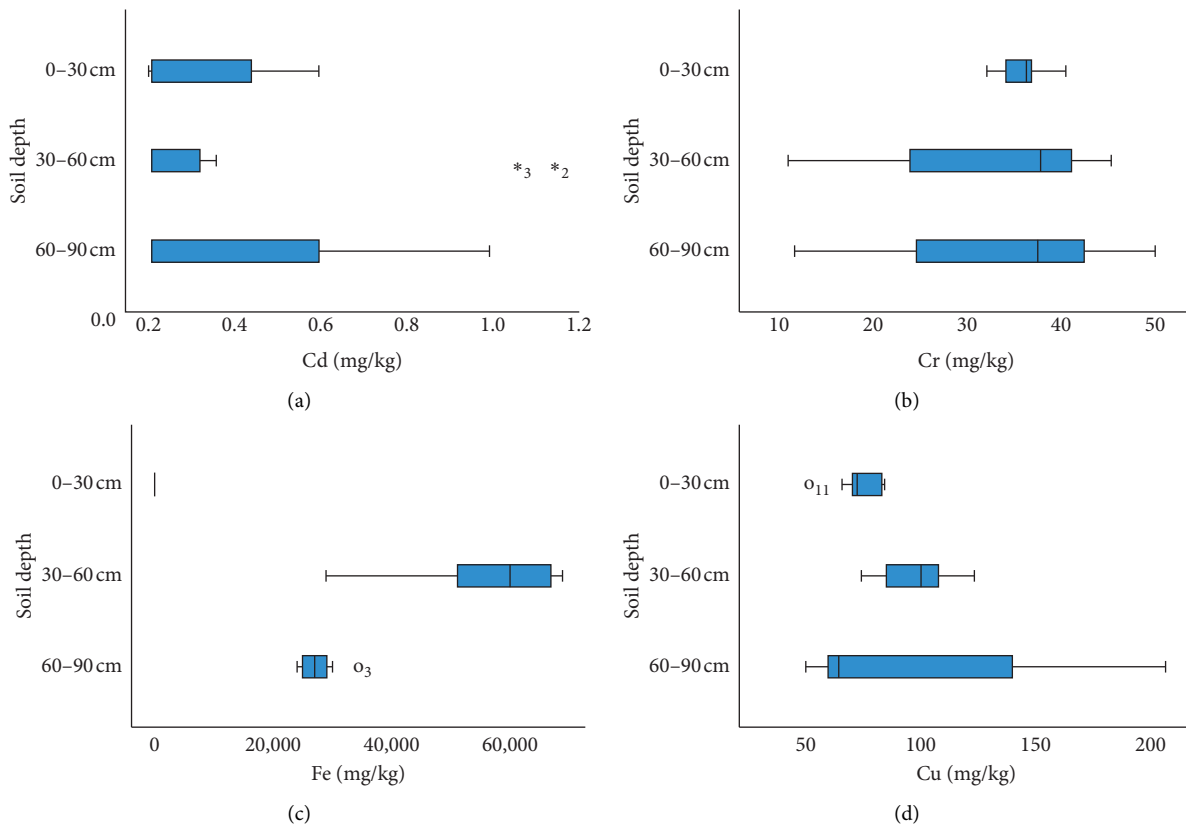


FIGURE 4: Continued.

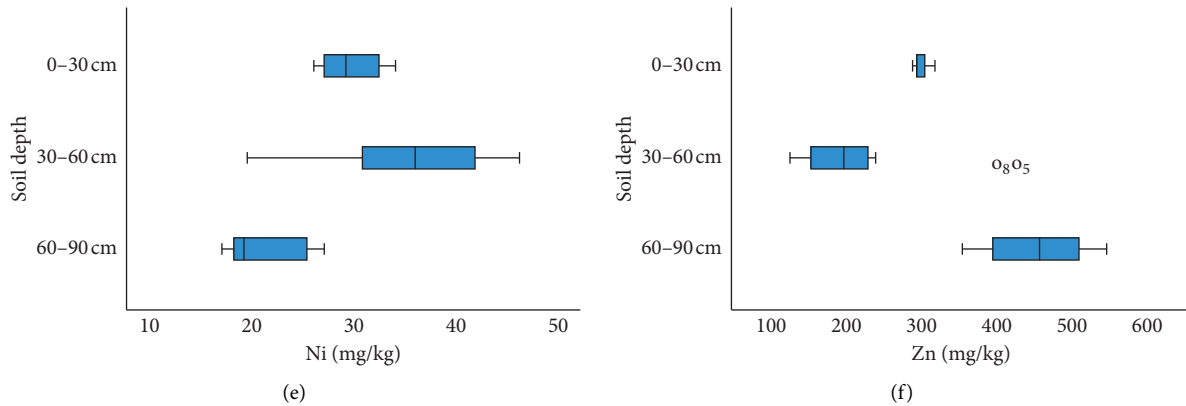


FIGURE 4: Variation of heavy metal concentration with depth around the landfill.

landfill. Higher concentrations of Cu in soils around the landfill compared to the control site could be an indication of the migration of leachate rich in Cu into the surrounding soils. Mean Cu concentrations increased to a depth of 30–60 cm but decreased further (Figure 4). The differences between soil depths were insignificant ( $p = 0.23$ ).

Iron content in soils at the Lumberstewart landfill was exceptionally high at all sites (Table 1). Significant differences were observed in Fe concentrations between soils from the landfill and those from the control site ( $p < 0.05$ ) (Table 1). Iron is contained in several household and industrial materials. It also occurs in high concentrations in some soils, and so, the high concentrations of Fe in the soils are not unexpected. Mean values for Fe concentration at a depth of 0–30 cm were significantly lower than those at a depth of 30–60 cm (Figure 4), which in turn were higher than Fe concentration values for soils at a depth of 60–90 cm ( $p = 0.001$ ).

Mean values for Ni concentration in soils from the landfill were higher than that of the control site (Table 1) ( $p < 0.05$ ). Nickel is a toxic element that is usually present in leachate generated in landfills with hazardous waste. It is derived from metal plating, electroplating, biosolids, and residue from the combustion of fossil fuels [64]. The presence of Ni in soils around the Lumberstewart landfill could indicate that the landfill received some industrial waste. There were significant differences in Ni concentration with depth ( $p = 0.001$ ), with mean values for soil Ni concentration at the different depths following the order 30–60 cm > 0–30 cm > 60–90 cm (Figure 4). There were no differences in Zn concentration between soils from the landfill environment and the control site (Table 1). However, some sites had Zn concentration values, which were lower than Zn concentrations in soils from the control site (Table 1). This could be a sign that soils in the area are generally rich in Zn. Significant differences were observed in the concentration of Zn in soils at different depths ( $p = 0.001$ ) as indicated in Figure 4.

The concentrations of the different heavy metals around the landfill are similar to what has been reported in soils around other municipal landfills [22, 23]. Though waste materials disposed in the landfill are the main sources of

heavy metals in landfill leachate and soils around the landfill environment, the covering and capping material could also contribute significant amounts of heavy metals. Fly ash is usually rich in heavy metals [65], and its use as a cover material in this landfill could have contributed to the observed heavy metal concentrations in leachate and eventually in the landfill environment. The pattern of mean Fe, Cu, Ni, clay content, and soil pH with depth around the landfill showed some similarities. Higher mean concentrations of Fe, Cu, and Ni at a depth of 30–60 cm compared to the other depths can be attributed to the depth of migration of leachate and to soil clay content and pH levels. Mean soil clay content and pH increased with depth, which might have encouraged the sorption and precipitation of these metals at this depth compared to lower depths. These elements have an affinity for organic matter and clay minerals and are precipitated at high pH levels [11–13]. Zinc, however, did not show any similarities with soil properties with depth. The patterns of heavy metal distribution with depth around the landfill environment can be attributed to the migration of leachate from the landfill and the soil properties.

**4.3. Relationship between Soil Properties and Heavy Metal Concentration.** Soil pH, texture, organic matter content, and cation exchange capacity influence the movement of heavy metals in soils [11, 12, 40]. Studies have shown that, in uncontaminated soils, heavy metal concentrations tend to display strong correlation with these soil properties. Chromium mobility, for example, is affected by soil pH and the amount of clay, Fe oxide, and organic matter in soils [66]. Binding of Cu to soil OM and clay minerals has been reported by Parkpian et al. [67]. Nickel is also reported to form covalent bonds with organic ligands, making OM relevant in its mobility in the soil environment. About 60% of Zn in both natural and uncontaminated soils is bound to the silicate lattice of the soil [67–69]. Pearson correlation analyses between heavy metals and the properties of soils at the landfill, however, showed weak correlation (Table 2). At a soil depth of 0–30 cm, significant correlations observed were Fe and clay/CEC, Zn and EC, Cd and OM, Cr and OM, and Cu and OM ( $p = 0.05$ ). Between a soil depth of 30 and



TABLE 2: Correlation coefficient between soil properties and heavy metals at different depths.

	Soil depth = 0–30 cm												
	Sand	Silt	Clay	pH	CEC	EC	OM	Fe	Ni	Zn	Cd	Cr	Cu
Fe	-0.46	0.34	0.58*	-0.02	0.75	0.05	-0.55	1					
Ni	-0.13	0.11	0.19	0.07	0.26	-0.29	0.32	0.16	1				
Zn	-0.40	0.48	0.18	0.01	0.21	-0.58	-0.03	0.29	0.39	1			
Cd	-0.23	0.17	0.29	-0.03	0.17	0.12	-0.68	0.36	-0.44	0.28	1		
Cr	-0.02	-0.03	0.19	0.28	0.52	0.52	-0.73	0.58	0.10	0.01	0.57	1	
Cu	-0.09	0.13	0.04	0.34	0.37	0.49	-0.65	0.36	-0.45	-0.22	0.66	0.71	1
	Soil depth = 30–60 cm												
Fe	-0.10	0.22	0.33	-0.53	0.44	-0.20	-0.58	1					
Ni	-0.15	0.53	0.53	-0.14	0.50	-0.29	-0.18	0.43	1				
Zn	0.47	-0.04	-0.36	0.35	-0.10	0.67	0.26	-0.73	-0.24	1			
Cd	-0.24	0.62	0.04	-0.63	0.64	0.20	-0.56	0.36	0.55	-0.04	1		
Cr	0.54	-0.67	-0.38	0.53	-0.55	0.18	0.18	-0.30	-0.69	0.15	-0.74	1	
Cu	0.28	0.27	-0.20	0.02	0.34	0.63	-0.17	-0.29	-0.02	0.74	0.21	-0.18	1
	Soil depth = 60–90 cm												
Fe	0.12	0.13	0.02	0.15	0.28	0.18	-0.37	1					
Ni	-0.16	0.42	0.58	-0.38	0.46	-0.23	0.02	-0.20	1				
Zn	-0.11	0.31	0.48	-0.37	0.35	-0.22	0.10	-0.32	0.96	1			
Cd	-0.12	0.09	0.35	-0.35	0.33	-0.35	-0.42	0.58	-0.07	-0.08	1		
Cr	0.75	-0.80	-0.63	-0.00	-0.66	0.36	-0.14	0.08	-0.40	-0.39	-0.14	1	
Cu	0.10	0.01	0.06	-0.13	0.30	0.06	-0.73	0.64	-0.24	-0.30	0.81	0.08	1

60 cm, Fe correlated with OM, Ni with clay, Zn with EC, and Cd, Cr, and Cu each with OM ( $p > 0.05$ ). Correlations observed at a depth of 60–90 cm were Ni and clay, Cr and clay/CEC, and Cu and OM ( $p > 0.05$ ). Most of these elements have an affinity for organic matter, which would have encouraged their sorption in the surface layers. However, this does not occur probably because the depth of heavy metal inputs in the landfill environment is beyond the surface layers of the soils. The randomness of the correlations between soil properties and heavy metals decreased with depth which could reflect natural pedogenic processes at lower depths of the soils. Unexplained correlation in the top 60 cm of the soils could be attributed to anthropogenic inputs of heavy metals into the soils.

**4.4. Heavy Metal Enrichment in Soils around the Closed Lumberstewart Landfill.** Except for Cd and Ni, the values of heavy metals in this study exceeded those of various standards (Table 1), indicating some level of pollution around the landfill. The extent of heavy metal pollution was further displayed by heavy metal CF values at the different sites (Supplementary Table 1). At a depth of 0–30 cm, CF values for Ni and Zn were less than two ( $CF < 2$ ), which indicates minimal contamination (Supplemental Table 1). Cadmium, Cr, Cu, and Fe recorded moderate enrichment ( $2 < CF < 5$ ), which shows moderate to significant contamination. The most enriched heavy metal at the surface of the soils around the landfill was Cu, followed by Cr then, Fe, Cd, Ni, and Zn. Heavy metal CF values for soils at a depth of 30–60 cm followed the order  $Cu = Fe > Cr > Cd > Ni > Zn$ , whereas at a depth of 60–90 cm, the order was  $Cr > Cu > Fe > Cd > Zn > Ni$  (Figure 5). Though Cd concentrations in the soils were very low, values for CF indicated

some additions of Cd into the soils around the landfill. This may be due to the high concentrations of Cd observed in some sites (Table 1). Except for Cr at a depth of 60–90 cm with a CF value that indicated very high contamination ( $CF > 6$ ), the level of contamination of the different metals could be described as mostly moderate to significant at a depth of 30–60 cm and moderate at depths of 0–30 cm and 60–90 cm, indicating significant enrichment between depths of 30 cm and 60 cm (Figure 5).

Pollution load index values for all sites around the landfill were above unity. According to Harikumar et al. [70], PLI values  $> 1$  implies that there is pollution, whereas PLI values  $< 1$  indicates no pollution. The most polluted sites around the landfill were site 6 at a depth of 0–30 cm, sites 2 and 3 at a depth of 30–60 cm, and site 7 at a depth of 60–90 cm. Pollution load index values at all sites indicated that the most contaminated depth was 30–60 cm (Figure 6), which was also the level with the most enrichment of heavy metals (Figure 5). This suggests that leachate migration took place or may still be taking place at this depth around the closed landfill. Migration of leachate at this depth could have occurred because of the textural properties of the soils. Mean clay content of the soils at this depth was relatively higher than what was obtained at a depth of 0–30 cm. Increase in clay content with depth would have restricted leachate infiltration to lower surfaces as a result of lower permeability and higher matric suction between the leachate and the soil particles, resulting in lateral rather than vertical migration of leachate within the 30 cm–60 cm depth around the landfill. The higher pH of soils at this depth could also have contributed to the higher concentrations of metals at this compared to other depths due to precipitation.

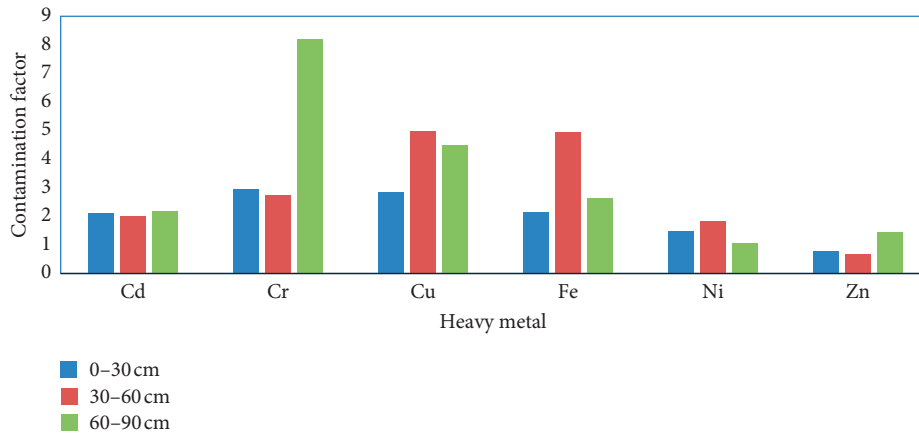


FIGURE 5: Heavy metal contamination in soils at different depths at the closed landfill.

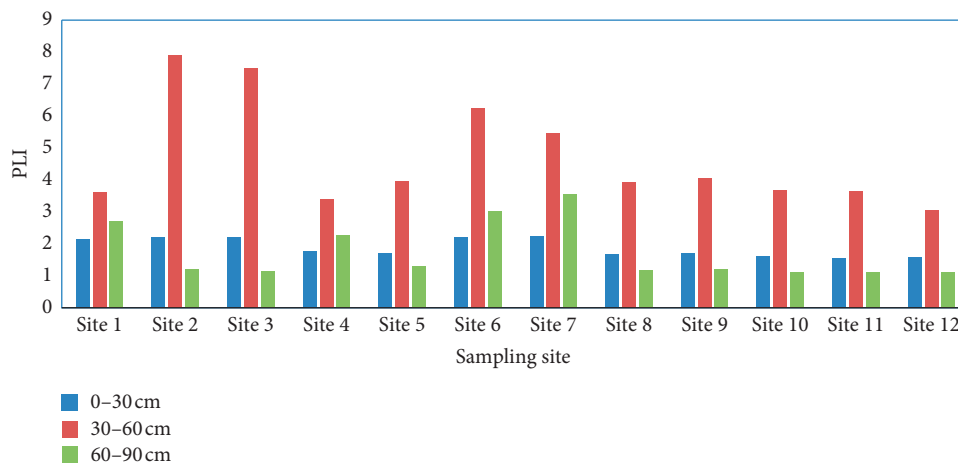


FIGURE 6: Pollution load index of the different sites around the landfill.

#### 4.5. Concentration of Heavy Metals in *Datura stramonium*.

Heavy metal concentration in *Datura stramonium* growing on soils around the landfill were higher than what was obtained in those growing at the control site (Figure 7). Heavy metals concentration in the plant followed the order  $Fe > Zn > Cu > Cr > Ni = Cd$ . Typical concentrations of heavy metals in this plant are not known, but comparison between samples from the control site and those obtained from the landfill indicate significant uptake in those growing around the landfill (Figure 7). Values for TF were, however, very low especially at the control site (Table 3). Highest TF values were obtained for Cd followed by Cu, then Zn and Cr, Ni, and Fe.

Plant samples having low heavy metal concentration (Cd) had higher TF values compared to plant samples having high metal concentration (Fe) which concur with previous findings by Loweshwari and Chandrappa [71] and Smith and Doran [72]. Ahmadipour et al. [11] have shown that heavy metals from anthropogenic sources are usually more available than those of natural origins. In this study, the correlation between CF and TF was very low. *R* values indicated that only 36.7%, 11.2%, 27.3%, 16.5%, 12.1%, and

62.4% of the variation observed in the transfer coefficient of Cd, Cr, Cu, Fe, Ni, and Zn, respectively, in *Datura stramonium* were accounted for by the extent of heavy metal enrichment in the soils.

Results from this study indicate that some transfer of heavy metals from soils to plants at the Lumberstewart landfill is going on. Though heavy metals TF values for *Datura stramonium* may be low, this may not be the case with other plants. Communities making use of this site for any kind of crop farming or ingesting the weeds present on the site for medicinal purposes might be exposed to heavy metals through prolonged ingestion. The health risk associated with exposure to heavy metals has been reported by Karimi et al. [25], Ngole-Jeme and Fantke [27], and Nicholls and Mal [73]. Though Fe is a necessary heavy metal in human physiology, high consumption of Fe may lead to DNA damage and loss of memory [74]. Epidemiological studies also associate high blood iron to insulin resistance and type 2 diabetes and cardiovascular disease [75]. These negative effects may be confounding with the toxicity of alkaloids that have been isolated in *Datura stramonium*. Surrounding communities making use of

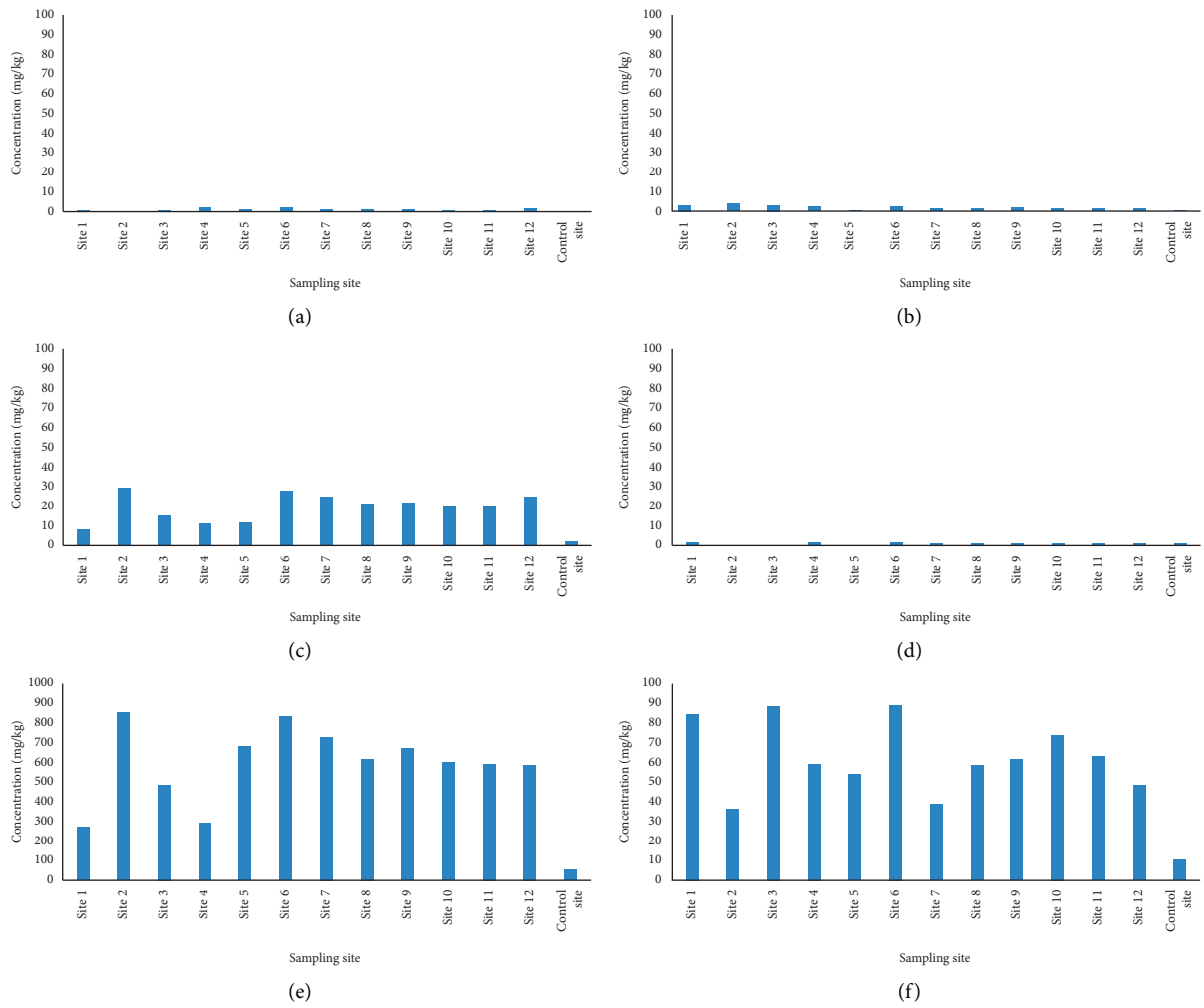


FIGURE 7: Concentration of heavy metals in Jimson weed. (a) Cd; (b) Cr; (c) Cu; (d) Ni; (e) Fe; and (f) Zn.

TABLE 3: Transfer coefficient of heavy metals in Jimson weed.

Site	Fe	Ni	Zn	Cd	Cr	Cu
Site 1	0.01	0.06	0.29	2.25	0.08	0.10
Site 2	0.03	0.00	0.12	0.03	0.09	0.42
Site 3	0.01	0.00	0.29	0.00	0.08	0.22
Site 4	0.01	0.06	0.19	19.55	0.06	0.16
Site 5	0.03	0.00	0.18	10.82	0.00	0.15
Site 6	0.03	0.05	0.30	6.19	0.06	0.34
Site 7	0.03	0.04	0.13	2.35	0.04	0.31
Site 8	0.03	0.05	0.20	10.45	0.04	0.29
Site 9	0.03	0.04	0.20	10.82	0.04	0.32
Site 10	0.03	0.04	0.25	9.55	0.04	0.31
Site 11	0.02	0.04	0.22	9.36	0.04	0.41
Site 12	0.02	0.05	0.16	19.00	0.04	0.37
Control site	0.00	0.06	0.03	0.00	0.00	0.08

*Datura stramonium* growing around this site may be at risk of exposure to heavy metals.

## 5. Conclusion

Closed landfill sites are used for various activities, but they are potential sources of heavy metals to the environment and could be a route of heavy metal exposure. Results from this study indicate that, several years after the closure of the Lumberstewart landfill, elevated concentrations of Fe, Cu, Ni, and Zn are still found in the soils especially at a depth of 30–60 cm. These metals show no correlation with the properties of the soils, which indicates nongeogenic origins. The contamination factor values indicated moderate to significant contamination at a depth of 30–60 cm and moderate contamination at depths of 0–30 cm and 60–90 cm. Values for PLI were higher at depths of between 30 and 60 cm, which were mostly above unity. Landfill site 6 was the most polluted at a depth of 0–30 cm, whereas at depths of 30–60 cm and 60–90 cm at sites 2 and 3 and site 7, respectively, were the most polluted. Heavy metal concentration in *Datura stramonium* followed the order Fe > Zn > Cu > Cr > Ni = Cd with highest concentrations commonly encountered at site 6 which also had the highest PLI values within the rooting depth of the plant. Heavy metal TF in the plants were highest for Cd and lowest for Fe. High TF values for Cd could be further investigated to identify any potential of the plant as a phytoremediator of Cd in contaminated soils. Though heavy metal TF values in *Datura stramonium* were low, continuous use of this weed by surrounding communities for medicinal purposes could result in heavy metal exposure and associated health risk. Continuous monitoring of the site needs to be ensured to mitigate the effects of continuous leachate migration from the closed landfill and to minimize its use by surrounding communities.

## Data Availability

The data used to support this study are available within the manuscript.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Supplementary Materials

This table presents details of the contamination factors and the pollution load indices of the different heavy metals in soils from the different sites. (*Supplementary Materials*)

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