CADMIUM, NICKEL, AND LEAD CONCENTRATION OF MUNICIPAL DUMPSITE IN WESTERN SAMAR, PHILIPPINES

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Received 21 September 2021/Accepted 8 March 2022

ABSTRACT

Heavy metal is one of the major problems due to its accumulation from the soil to the food chain, wherein dumpsites are the primary sources of heavy metal pollution. This study aimed to determine the presence of heavy metals in the soil of Santa Rita, Western Samar dumpsite and to quantify them to obtain knowledge on the possible high contamination that may affect the surrounding areas. This study focused on the presence and concentrations of heavy metals Cd, Ni, and Pb. Eighteen (18) soil samples were acquired within the three sampling sites: shoulder slope, main dumpsite, and foot slope. Each sampling site has three sampling points with a depth of 0 - 30 cm and 30 - 60 cm. Analysis showed that all heavy metals are present in the dumpsite, and the concentrations ranged from 0 - 0.1 mg/kg, 0.09 - 3.7 mg/kg, and 0.09 - 3.7 mg/kg for cadmium, lead, and nickel, respectively. In comparing heavy metals within the sampling sites and depths, only cadmium has a significant difference, while Ni and Pb have no significant difference. Compared with WHO standards, all heavy metals tested still fall within the standard limit. Therefore, the dumpsite is still at a safe level. However, residents should take measures to maintain the soil quality since heavy metal contamination in dumpsites is likely to exacerbate.

Keywords: cadmium, lead, municipal dumpsite, nickel, soil pollution

INTRODUCTION

One of the global issues is improper waste management. Open dumping and open burnings are the most implemented waste treatment and final disposal systems, mainly available in low-income countries (Ferronato & Torretta 2019). The increased population and the rising demand for food and other essentials equate to increased waste generated daily in each household, wherein this waste eventually dumps in dumpsites and impacts the soil and the surface environment (Mekonnen *et al.* 2020). Uncontrolled disposal results in severe heavy metal contamination (Vongdala *et al.* 2019). Further, dumpsite operation is a source of heavy metal pollution that can affect the biosphere (Sakawi *et al.* 2013).

Heavy metals naturally occur in the earth's crust with high atomic weight and density at least five times greater than water (Tchounwou *et al.* 2012). Heavy metal is one environmental contaminant affecting aquatic and terrestrial ecosystems (Sakawi *et al.* 2013). Heavy metals possess a severe risk because they tend to bioaccumulate, which means an increase in chemical concentration in a biological organism over time compared to the chemical concentration in the environment (Helmenstine 2021). Sources of heavy metals include mining and plating, fertilizers and pesticides, sludge dumping, and municipal waste (Mudgal 2010). The liquid that exudes and percolates solid wastes eventually transfers to the soil. Heavy metals, pesticides, and hydrocarbons are threatening substances that bind in liquid and constantly contaminate soil and water (Adelekan & Alawode 2011).

Dumpsite of Santa Rita is located at Sitio Canonay of Barangay Rosal, Santa Rita, Western Samar. The dumpsite is already 20 years old, and all kinds of waste are accepted from all

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barangays of Santa Rita, Western Samar. The municipal government of Santa Rita owns the entire area of the dumpsite. The main dumpsite is 45 m from the nearby residences and 22 m away from the rice plants in Sitio Canonay.

This study aimed to determine and quantify the concentrations of heavy metals present in the Santa Rita, Western Samar dumpsite soil and provide knowledge about the possible high contamination of heavy metals that may affect the surrounding areas.

MATERIALS AND METHODS

Study Site

The study was conducted in Santa Rita, Western Samar, where the dumpsite is located at Sitio Canonay of Barangay Rosal (Fig. 1). The dumpsite was divided into three sampling sites: shoulder slope, main dumpsite, and foot slope, wherein three sampling points in each sampling site were randomly selected. The dumpsite comprises verdant hills covered with grassland and forest vegetation on the shoulder slope, a bulk of wastes that covers the main dumpsite with *Ipomoea aquatica* (water spinach) plantation that surrounds some parts of the area, and a rice field that could be found at the foot slope of the dumpsite.

Sample Collection, Preparation, and Heavy Metal Analysis

A descriptive analysis of research was utilized in this study. Each sampling point was dug up to at least 60 cm depth. Then, two (2) composite samples in every sampling point were collected according to their depth (0 - 30 cm and 30 - 60 cm), wherein 18 soil samples were acquired within the three sampling sites. After sampling, all soil samples were air-dried, crushed using a mortar and pestle, sieved through a 2-mm sieve, and stored in a clean, resealable, properly labeled bag.

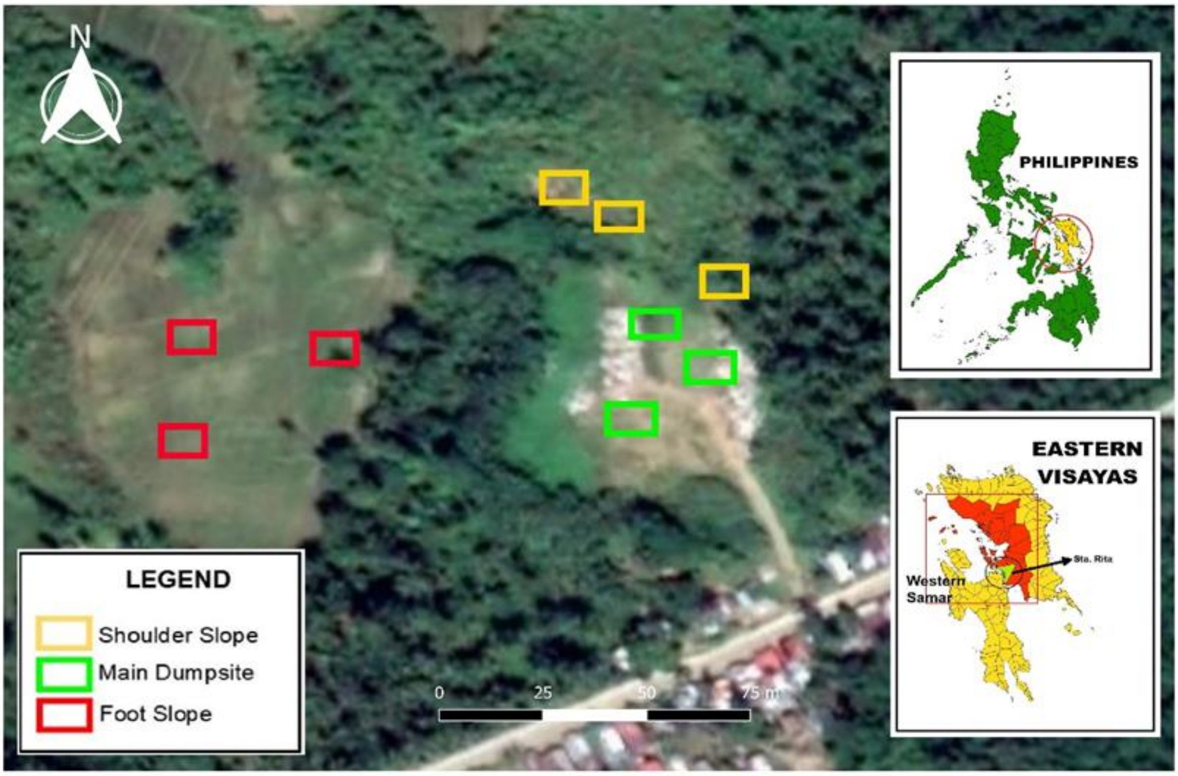

Figure 1 Location map showing the sampling points

Soil samples were digested using the DTPA Acid extraction solution. The digested soil samples' Cd, Ni, and Pb concentrations were analyzed using Atomic Absorption Spectrophotometry (AAS) equipped with Pb hollow cathode lamp (Varian SpectraAA 220 FS with SIPS pump unit and autosampler SPS-5). Standard solutions of the heavy metals were utilized in the preparation of the calibration curve. The following formula was used to compute Pb, Cd, and Ni concentration in the dumpsite soil.

$$\text{Heavy metals in soil (ppm)} = \frac{\text{mL of extraction solution}}{\text{grams of soil}} \text{ x conc. (ppm)}$$

where:
mL of extraction solution = volume of extraction solution
grams of soil = weight of soil sample in grams
conc. (ppm) = result of AAS concentration in mg/L

Statistical Analysis

Two-way analysis of variance (ANOVA) was used in this study to determine the significant variation in the mean concentrations of identified heavy metals in soil concerning sampling depth and sampling site. Natural Logarithm was used to obtain the assumption of ANOVA in which all data should be in a normal distribution and have homogenous variance. Shapiro Wilk's test was used for analyzing the normality, and Levene's test was used to determine the homogeneity. In comparing the concentrations of identified heavy metals with standard, a two-tailed t-test was used in this study. All statistical computation was run in the program R Project where in to run the statistical computing in R software, packages used were dplyr, car, rstatix, tidyverse, and ggpubr (R Core Team 2021). Acceptance and rejection of the hypothesis were set at a 0.05 level of significance.

RESULTS AND DISCUSSION

Heavy Metal Contents of Dumpsite Soil

Cd, Ni, and Pb in different sampling sites at different depths were found in the study site (Table 1). The environment and its components in Santa Rita have been seriously polluted by heavy metals, which have compromised the ability of the environment to foster life and take its intrinsic values. Heavy metals commonly occur naturally on earth, but anthropogenic activities result in large quantities of different environmental components (Masindi & Muedi 2018). The presence of nickel could be attributed since nickel is a metal with widespread distribution in the environment, including minerals. It is an essential constituent with many industrial and commercial uses (Iyaka 2011). Nickel can exist in soils in several forms, such as inorganic crystalline minerals or precipitates, complexed or adsorbed on organic cation surfaces, or inorganic cation exchange surfaces, among others (Cempel & Nikel 2006). Nickel is a ubiquitous pollutant and carcinogen toxic metal found in many hazardous waste sites, especially in dumpsite that receives different types of waste, such as municipal solid waste, clinical waste, and industrial waste. Industrial waste materials, lime, fertilizer, and sewage sludge contribute to the significant nickel sources in the soil (Das *et al.* 2018).

Table 1 Concentrations of heavy metals in dumpsite soil at different sampling sites and depths

Sampling Sites	Sampling Points	GPS Coordinates	Cd (mg/kg)		Ni (mg/kg)		Pb (mg/kg)	
			0 - 30 cm	30 - 60 cm	0 - 30 cm	30 - 60 cm	0 - 30 cm	30 - 60 cm
Shoulder Slope	Point 1	N 11°28'9.52364" E 124°57'7.20043"	Trace	Trace	3.717	3.054	0.246	0.327
	Point 2	N 11°28'10.09024" E 124°57'6.34467"	Trace	Trace	0.494	0.378	Trace	Trace
	Point 3	N 11°28'10.32682" E 124°57'5.89446"	Trace	Trace	2.715	3.679	0.795	0.566
	Mean		**-**	**-**	**2.308667**	**2.370333**	**0.5205**	**0.4465**
Main Dumpsite	Point 1	N 11°28'9.17942" E 124°57'6.63555"	0.0114	0.1094	0.446	1.078	1.51	58.642
	Point 2	N 11°28'8.81745" E 124°57'7.07731"	0.0014	Trace	2.154	1.91	0.318	0.846
	Point 3	N 11°28'8.4058" E 124°57'6.42071"	Trace	0.0504	1.939	1.065	0.7	11.578
	Mean		**0.0064**	**0.0799**	**1.513**	**1.351**	**0.842667**	**23.68867**
Foot Slope	Point 1	N 11°28'9.05403" E 124°57'2.92525"	0.0024	0.001	0.167	0.175	2.291	1.893
	Point 2	N 11°28'8.94994" E 124°57'4.08035"	Trace	0.001	0.448	0.409	1.438	1.525
	Point 3	N 11°28'8.17159" E 124°57'12.86853"	Trace	0.0023	0.085	0.147	1.469	1.56
	Mean		**0.0024**	**0.001433**	**0.233333**	**0.243667**	**1.732667**	**1.659333**

On the other hand, lead was found at all sampling sites and depths within the dumpsite of Santa Rita, Western Samar. Lead could be found in the dumpsite because it has some unique physical and chemical properties from historical times and has become a common environmental pollutant. Lead forms different complexes with soil elements. It only takes a small piece of the lead as these complexes within the soil are phytoavailable (Pourrut *et al.* 2011). Lead has remained in the ground for thousands of years, not biodegradable (EPA 2019). It poses a severe environmental risk due to its continued usage in every part of the world due to the abundance of gasoline, industrial processes, lead-based painting, lead-containing pipes, and lead-acid batteries (Wani *et al.* 2015). Lead consumption has increased due to increased demand for lead-containing products, such as batteries, electronic products, cathode ray tubes, PVC stabilizers, and lead pigments. These lead-containing products increase lead-containing waste (European Commission DG ENV 2002).

Moreover, cadmium was also found within the dumpsite of Santa Rita. Cadmium can be released into the environment through anthropogenic activities and can be used in many industrial uses, such as plastic, pigment, enamels, ceramics, and steel plating. In addition, industrial processes produce cadmium as a by-product (Hasan *et al.* 2019).

Results of our study showed that nickel is found at all sampling points and depths. Lead was present in trace amounts at the shoulder slope point 2 and other sampling points and depths. Cadmium was mainly found at the foot slope at 30 - 60 cm depth, sparsely found at the main dumpsite, and was present only in trace amounts at the shoulder slope. The mean concentrations of cadmium, nickel, and lead in soil ranged from 0 - 0.1 mg/kg, 0.09 - 3.7 mg/kg, and 0.09 - 3.7 mg/kg for cadmium, nickel, and lead, respectively. Mean concentrations were 0.0084±0.0030 mg/kg, 1.3517±0.4382 mg/kg, and 1.0959±0.2475 mg/kg, respectively, for 0 - 30 cm depth and 0.0137±0.0122 mg/kg, 1.3217±0.4322 mg/kg, and 1.1195±0.2560 mg/kg respectively for 30 - 60 cm depth.

The results agree with the result from Orodu *et al.* (2017), who reported a range of 0.09 - 1.10 mg/kg, 0.19 - 1.84 mg/kg, and 0.47 - 14.33 mg/kg for cadmium, nickel, and lead, respectively with means of 0.22±0.08 mg/kg, 1.03±0.16 mg/kg, and 5.17±5.04 mg/kg. Results of our study also agree with Amos-Tautua *et al.* (2014), which determines the concentration of heavy metals in the dumpsite and cadmium. They reported a range of < 0.000.1±0.01 mg/kg, while Amadi *et al.* (2012) studied different heavy metals, cadmium, and lead, resulting from a mean concentration of 0.15 ppm and 16.0 ppm, respectively. Essien *et al.* (2019) reported a range of 0.92 - 1.4 mg/kg, 1.8 - 3.7 mg/kg, and 0.79 - 0.98 mg/kg taken during the wet season while 0.124 - 1.4 mg/kg, 1.72 - 2.98 mg/kg and 0.94 - 4.12 mg/kg taken during the dry season for nickel, lead and cadmium, respectively. It can be seen that heavy metal concentrations followed the order Ni > Pb > Cd, which is in agreement with the results from Orodu *et al.* (2017), Amadi *et al.* (2012), Essien *et al.* (2019), and Makuleke & Ngole-jeme (2020).

Figure 2 shows the mean concentrations of the metals from each sampling site at different depths. The trends observed at a sampling depth of 0 - 30 cm were generally the same as those taken at 30 - 60 cm. Differences in heavy metal concentration can be attributed to the amount of waste in the dumpsite carrying the different heavy metals. For example, nickel having the highest levels could be attributed to more nickel-containing wastes being dumped, such as buttons, zips, coins, household appliance tools, and other consumer products (Asemave & Annwange 2013). Extensive industrial use of nickel has led to widespread environmental pollution (Das *et al.* 2018).

On the other hand, cadmium generally has the lowest concentration. This could be due to the decrease in demand for cadmium-containing products in the industry. Specifically, the use of cadmium for pigments, PVC stabilizers, and plating has been phased out. As an impurity in zinc and fertilizers, turnover of cadmium has also been significantly decreased due to refining and changes in raw materials (European Commission DG ENV 2002), resulting in a decrease in cadmium-containing products that end up in waste.

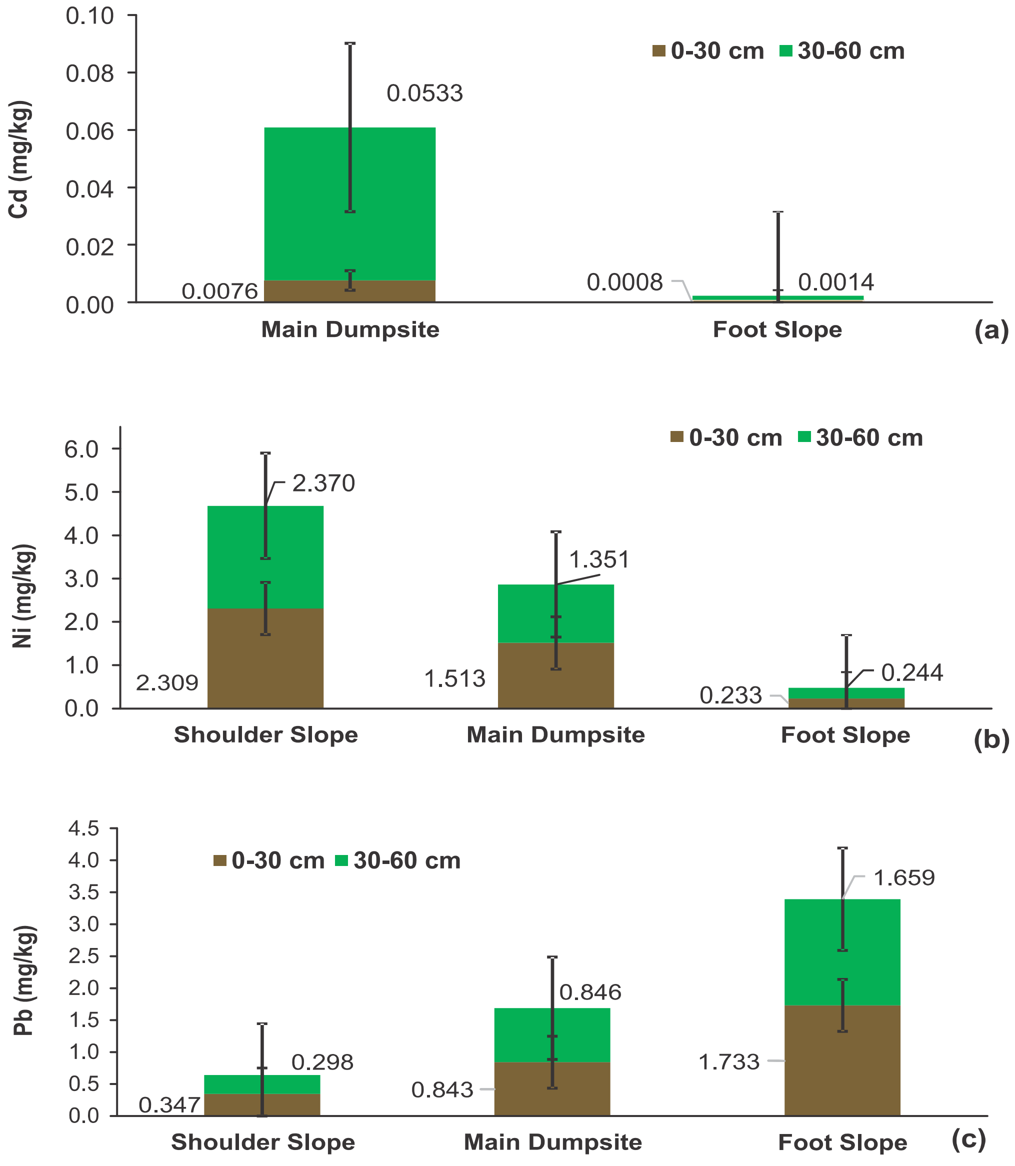

Figure 2 Comparison of (a) Cd, (b) Ni, and (c) Pb concentration means per sampling site and depth

Comparison of Heavy Metal Contents According to Sampling Site and Depth

Table 2 presents the statistical analysis results of comparing concentrations of cadmium, nickel, and lead at the different sampling sites and depths. Significant differences are indicated by $P \leq 0.05$. In case of significant differences, the results of post-hoc Tukey's Honest Significant Difference (HSD) are indicated in Table 3.

The mean values of Cd, Ni, and Pb for all sampling sites were 0.008 mg/kg, 1.351 mg/kg, and 1.096 mg/kg, respectively, for 0 - 30 cm depth. While for 30 - 60 cm depth, the mean values of Cd, Ni, and Pb were 0.033 mg/kg, 1.322 mg/kg and 1.119 mg/kg, respectively for all sampling sites (Table 2). The computed P values showed no significant differences between the two sampling depths for all heavy metals tested.

In addition, the computed P values of Cd, Ni, and Pb in each sampling site for both sampling depths are 0.001, 0.003, and 0.012, respectively. It means that all sampling sites differed and suggested that dumpsite soil lacks the capacity to impede the downward migration of leachate from topsoil to subsoil (Amadi *et al.* 2012). Moreover, only cadmium had a depth-site interaction wherein two independent variables should be tested whether the effect of one independent variable is dependent or affects the other.

Table 2 Differences in cadmium, nickel and lead concentrations between different sampling sites and depths

Sampling depth				
Heavy metals	Mean concentration (mg/kg)		P value (α = 0.05)	Interpretation
	0 - 30 cm	30 - 60 cm		
Cd	0.0084	0.03282	0.9421	Not significant
Ni	1.3517	1.32166	0.8945	Not significant
Pb	1.095875	1.1195	0.9320	Not significant

Sampling site					
Heavy metals	Mean concentration (mg/kg)			P value (α = 0.05)	Interpretation
	Shoulder slope	Main dumpsite	Foot slope		
Cd	-	0.04565	0.001675	0.0006	Significant
Ni	2.3395	1.432	0.2385	0.0026	Significant
Pb	0.4835	0.8435	1.13067	0.0115	Significant

Depth - Sampling Site Interaction		
Heavy metals	P value (α = 0.05)	Interpretation
Cd	0.0207	Significant
Ni	0.9767	Not significant
Pb	0.9505	Not significant

Table 3 presents results for multiple pairwise comparisons using Tukey's Honest Significant Difference test for the indicated heavy metals. This is employed to identify which groups are significantly different from each other.

Results was found that cadmium levels were approximately 3 times higher in the main dumpsite than in the foot slope. For nickel and lead, only the main effect of the sampling site was found to be significant. Nickel concentrations at both the main dumpsite and shoulder slope are approximately 2 times higher than concentrations at the foot slope (Table 3). This suggested a stronger upward lateral leaching (Raji & Adeoye 2017), leading to more nickel accumulation at the shoulder slope than at the foot slope. In the case of lead, levels at the foot slope were found to be 1.3 times higher compared to the shoulder slope, suggesting a slightly higher downward lateral leaching (Raji & Adeoye 2017). Differences in leaching strength can be attributed to differences in soil properties (Asemave & Anhwange 2013; Makuleke & Ngole-jeme 2020).

To better visualize the interaction between the depth effect and sampling site effect for cadmium, an interaction plot is presented in Figure 3. An interaction effect is the simultaneous effect of two or more independent variables on one dependent variable. Their combined effect is significantly greater (or substantially less) than the sum of the parts. The presence of interaction effects in survey research is essential because it tells researchers how two or more independent variables impact the dependent variable.

Table 3 Tukey's HSD results for nickel, lead, and cadmium

Heavy metal	Sampling site	Difference	P value (α = 0.05)	Interpretation
Ni	Main Dumpsite - Foot Slope	1.8365680	0.0092866	Significant
	Shoulder Slope - Foot Slope	2. 1108135	0.0035699	Significant
Pb	Shoulder Slope - Foot Slope	-1.3433094	0.0104619	Significant
	Main Dumpsite - Foot Slope	2.9373	0.0006842	Significant
Cd	Depth - Sampling Site Interaction	Difference	P value (α = 0.05)	Interpretation
	60 cm Main Dumpsite - 30 cm Foot Slope	3.4320323	0.0103995	Significant
	30 cm Main Dumpsite - 60 cm Foot Slope	2.1559770	0.0191866	Significant
	60 cm Main Dumpsite - 60 cm Foot Slope	4.0298647	0.0018821	Significant
	60 cm Main Dumpsite - 30 cm Main Dumpsite	1.8738877	0.0421485	Significant

Cadmium concentration at the main dumpsite is 3 times higher than the foot slope (Fig. 3). However, differences can be observed due to the interaction between depth and sampling site effects. Specifically, cadmium concentration at the main dumpsite (0 - 30 cm) is 2 times higher compared to the foot slope (30 - 60 cm), 3 times higher at the main dumpsite (30 - 60 cm) than the foot slope (0 - 30 cm), and 4 times higher at the main dumpsite (30 - 60 cm) compared to foot slope (30 - 60 cm).

Comparison of Heavy Metal Contents with Standard Permissible Limits in Soil

Table 4 presents the results for comparison of heavy metal concentrations with the maximum permissible limits set by the World Health Organization (WHO). The permissible limits of heavy metals in soil are 0.02 - 0.5 (mg/kg), 0.1 - 5 mg/kg, and 0.3 - 10 mg/kg for cadmium, nickel, and lead, respectively (WHO 2001). A soil sample is polluted with a particular heavy metal if the concentration exceeds the upper limit of the acceptable range (Ogundele *et al.* 2015). Therefore, to determine whether soil samples from Santa Rita, Western Samar dumpsite was polluted with the indicated heavy metals, the right-tailed t-test was employed to compare the sample's mean concentration with the maximum permissible.

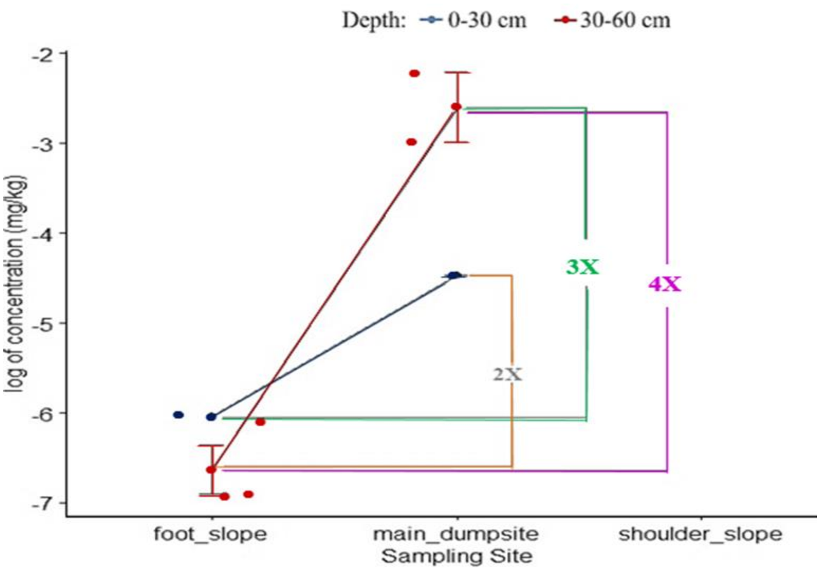

Figure 3 Depth - Sampling Site interaction plot for cadmium

Table 4 t-test results for comparison of mean concentrations with WHO standard

Heavy metals	Sampling sites	Sampling depth (cm)	Mean concentration	WHO standard	P-value (α = 0.05)	Interpretation
Cd	Shoulder Slope	0 - 30	N/A	0.05	N/A	N/A
		30 - 60	N/A	0.05	N/A	N/A
	Main Dumpsite	0 - 30	N/A	0.05	N/A	N/A
		30 - 60	0.0799	0.05	0.2479	No significant difference
	Foot slope	0 - 30	N/A	0.05	N/A	N/A
		30 - 60	0.0014	0.05	1	No significant difference
Ni	Shoulder Slope	0 - 30	2.3087	5	0.9471	No significant difference
		30 - 60	2.3703	5	0.9391	No significant difference
	Main Dumpsite	0 - 30	1.513	5	0.9885	No significant difference
		30 - 60	1.351	5	0.9971	No significant difference
	Foot slope	0 - 30	0.2333	5	0.9997	No significant difference
		30 - 60	0.2437	5	0.9998	No significant difference
Pb	Shoulder Slope	0 - 30	0.5205	10	0.9908	No significant difference
		30 - 60	0.4465	10	0.9960	No significant difference
	Main Dumpsite	0 - 30	0.8427	10	0.9993	No significant difference
		30 - 60	23.689	10	0.2606	No significant difference
	Foot slope	0 - 30	1.7327	10	0.9994	No significant difference
		30 - 60	1.6593	10	0.9999	No significant difference

The results obtained showed that all heavy metal concentrations were not significantly higher than the maximum permissible limits set by the WHO (Table 4). As such, they still fall within the permissible limits of WHO. It means that the concentrations of all indicated heavy metals are still at safe levels. Heavy metals in the dumpsite soil should not threaten anyone, particularly the surrounding susceptible environment. However, serious measures should still be taken to maintain soil quality since heavy metal contamination in dumpsites will likely exacerbate in the future (Orodu *et al.* 2017).

CONCLUSION

Identified heavy metals are all found at different sampling sites and depths of Santa Rita Western Samar dumpsite soil. The mean concentrations (mg/kg) of Ni is the predominant, followed by Pb, and the least is Cd. No significant differences were observed in the concentrations of Pb and Ni in different sampling sites and depths, but significantly different in the concentration of Cd. In addition, no significant differences were observed between the concentrations of identified heavy metals in all sampling sites at different depths and the standard permissible limits. Therefore, all concentrations of identified heavy metals were within the permissible limits in soil.

ACKNOWLEDGMENTS

The authors acknowledge Prof Fe T. Piedad, Prof Adorne Y. Madera, and Prof Grechelle N. Socias for their essential contributions during the final polishing of this manuscript.

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