

The Levels of Heavy Metals in the Soil of Illegal Open Dumpsites in Malaysia

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ABSTRACT: This study aims to understand the soil impacts of illegal dumpsites in Malaysia through quantifying the heavy metals in the soil of two dumpsites, one receiving construction waste and the other receiving municipal solid waste. Five soil samples were collected from each dumpsite, and sampling was repeated in the second week to examine the temporal changes in the levels of heavy metals. All sampling was conducted in triplicates. The soil samples were sieved, dried, and digested with aqua regia at 70 °C, after which the digested mixtures were filtered. The filtrates were diluted and tested with an atomic absorption spectrophotometer for heavy metals. The soil heavy metal concentration ranges were as follows: Al (24.67-142.20 mg/kg), Cd (< 0.01-0.083 mg/kg), Cu (0.10-14.99 mg/kg), Fe (11.20-241.77 mg/kg), Mn (0.09-22.60 mg/kg), Ni (0.02- 0.77 mg/kg), and Zn (0.14-35.03 mg/kg). All the heavy metals have been detected at all the sampling points except that the Cd levels at some sampling points were below the detection limit. The levels of heavy metals varied spatially and temporally, though higher Cd, Cu, Fe, Mn, Ni, and Zn were detected consistently at two sampling points of the dumpsite receiving municipal waste. This could be linked to the electrical and electronic waste at the dumpsite. The levels of heavy metals in the soil did not constitute soil contamination. However, it is important to control illegal dumping activities to reduce the associated health and safety concerns, such as infestation of vermin, fire, physical hazards, and odor.

KEYWORDS: Illegal dumpsites; heavy metals; soil; contamination; construction waste; municipal waste

1. Introduction

The release of heavy metals into the environment is an environmental issue affecting the health of living organisms on Earth [1]. Heavy metals comprise metalloids and metals which usually have atomic densities larger than 5.0 g/cm³ [2]. Heavy metals are poisonous and toxic, even at relatively low concentrations [3]. Heavy metal contamination affects abiotic systems, including the air, soil, and water that living organisms depend on for survival. Heavy metals have been reported to produce carcinogenic, teratogenic, mutagenic, and neurotoxic effects on living

organisms [4]. Thus, the cumulative long-term health effects of heavy metal contamination have become a leading concern throughout the world [3].

Anthropogenic activities, particularly agriculture and waste disposal, are reported to be two of the main causes of soil contamination with heavy metals [4]. Soil contamination with heavy metals is more severe in developing countries where the efficiency of municipal solid waste (MSW) management, the coverage of MSW collection, and the recycling rates of MSW are relatively low [5]. In developing countries, a lack of recycling technology for MSW results in higher recycling costs and a proclivity to landfill MSW [6]. With lower coverage of MSW collection, much of the waste is often dumped illegally in open spaces, resulting in open dumpsites. In Malaysia, the generation of MSW has increased with the increasing population, reaching approximately 38,427 tonnes per day as of 2021, and 82.5% of the waste ends up in landfills. However, the recycling rate for 2020 was only 30.7%, significantly lower than the 50% of its developed neighbor, Singapore [7]. Landfills in Malaysia are facing the mounting pressure of increasing MSW loads, hitting 14 million tonnes per year by 2022, and there is increasing concern that much of the waste is not making its way to proper landfill facilities [7]. A study revealed that there were 64 illegal dumpsites for construction waste in a single district alone, accommodating an approximate total of 427.3 m³ of the waste [8]. Since this is the case, it seems likely that there are more illegal dumps in the district that take in both MSW and construction waste.

The large number of unaccounted open dumpsites throughout the country has humongous implications for the quality of the soil surrounding the dumpsites. While MSW is properly contained and the leachate is adequately managed in proper landfill facilities, illegal dumpsites lack these pollution control mechanisms to minimize the contamination caused by the MSW therein to their surroundings [9]. Even with some level of management, it has been reported that heavy metals such as Mn, Fe, Ni, Cr, Pb, Zn, Co, and Cu were detected in the soil horizons beneath the waste disposal zone of a landfill [10]. The concentrations of the heavy metals were higher at the soil-waste interface than at the up-slope area of the waste disposal zone. They were also found to be higher at the surface soil layer and to decrease with increasing soil depth [10]. Another study conducted for two different MSW disposal sites in Malaysia, namely the Panchang Bedena Landfill and the Kelana Jaya ex-disposal site, revealed that the former had levels of heavy metals within the Dutch Intervention Value for Soil Remediation while the latter had arsenic and mercury levels exceeding the intervention values [11]. This implies the importance of adequate management of waste disposal sites to reduce their environmental impacts. A study probing the distribution of heavy metals in a non-sanitary landfill (Sungai Kembong) and three sanitary landfills (Air Hitam, Kubang Badak, and Ampar Tenang) found higher concentrations of heavy metals at deeper soil layers than the upper soil, in contrast to the higher levels reported at the soil-waste interface and surface soil layer [11], thus indicating that downward migration of heavy metals seems to have occurred over time. A study examining the soil contamination at the dumpsite of the bottom ash generated from hospital waste incineration showed that concentrations of heavy metals exceeded the permission limits with Zn recording the highest concentration (16417.69 mg/kg), followed by Pb (143.80 mg/kg), Cr (99.30 mg/kg) and Cd (7.54 mg/kg) [12]. These studies, though conducted in different settings of waste disposal sites, pointed to the concern of soil contamination with heavy metals at and around these sites.

The reason that MSW is usually linked to heavy metals is that certain wastes contain heavy metals inherently. Batteries, for instance, are known to be a significant source of lead and cadmium [12]. Plastics, lead crystal glass, and solders are also important sources of lead. As for mercury, it could come from dental amalgam as well as measurement and control devices such as thermometers [13]. It is likely that heavy metals could leach from MSW, especially in an open dumpsite where the MSW is exposed to the elements and there is no lining to prevent the infiltration of leachate into soil, let alone a leachate collection and treatment system to render the leachate harmless [14]. At such sites, lateral and vertical movements of contaminants could occur, leading to a larger area of contamination than the zones where the MSW is placed. There are multiple regional studies tracing the levels of heavy metals in soil around dumpsites. For instance, in Nigeria, Ogunbanjo et al. studied the forms of heavy metals in the soil at two dumpsites and disclosed substantial pollution of the soil by Cd and Cu, leading to an unacceptable cancer risk [15]. Another study conducted for an MSW dumpsite located in Ghana showed the contamination levels for the dumpsite, down site, and top site in descending sequence with a high level of Fe recorded [16]. These studies pointed to regional variations in the soil impacts of dumpsites, hence their regional significance. While studies on soil contamination with heavy metals due to landfills and dumpsites have been conducted in Malaysia, there is currently a lack of such studies in the Malaysian Borneo [9][10]. This study therefore aims to fill in this regional gap and provide incremental insight into the extent of soil pollution caused by illegal dumpsites. It aims to quantify the concentrations of heavy metals in the soil in and surrounding two open dumpsites located in Miri on Borneo Island. It then evaluates if the concentrations of the heavy metals are acceptable in reference to the established guideline values. As previous studies have shown, heavy metals in soil move around over time. This study also looks at how the concentrations of heavy metals in soil change over a two-week period.

2. Materials and Methods

A total of 5 sampling points were identified within and around two dumpsites. The locations of the dumpsites and the respective sampling points are shown in Figures 1 and 2. The GPS coordinates of the sampling points are provided in Table 1. Sampling was conducted twice, with the second two weeks apart from the first to examine the temporal changes in the concentrations of heavy metals. This study presents a preliminary attempt to track the short-term temporal variations in the levels of soil heavy metals, and the second sampling was conducted two weeks after the first, assuming that significant changes in the levels might have occurred over the duration. Besides, sampling intervals of two weeks and one month were commonly reported [17]. During sampling, a hand auger was used to obtain a soil column at each sampling site. Soil at a depth of 15 cm was collected and transferred to a labelled plastic bag to be brought back to the laboratory for storage and analysis. Ten samples were collected during the initial sampling and another 10 during the second sampling, yielding a total of 20 soil samples.



Figure 1. The location of Open Dumpsite A (a) and the sampling points (b).

Each soil sample was gently homogenized and sieved through a 2 mm sieve. The sieved sample was dried in an oven at a temperature of 40 °C for approximately 30 minutes. 1 g of the dried sample was placed in a 250 ml beaker. 15 ml of aqua regia were prepared by mixing 35% of HCl (3 ml) and 70% of high purity HNO₃ (9 ml). The aqua regia was added to the 250 ml beaker containing 1 g of sample. The mixture was digested using the Titan Microwave Sample Preparation System at 70 °C. The digested mixture was filtered using a filter paper, and the filtrate was transferred to a 100 ml volumetric flask. The solution was subsequently diluted by adding deionized water to the marked level of the volumetric flask. The solution was analyzed for the concentrations of Al, Cd, Mn, Cu, Fe, Ni, and Zn using an atomic absorption spectrophotometer (AAS). A triplicate of each soil sample was prepared for analysis. Prior to the analysis, a calibration curve for each heavy metal of interest had been constructed with standard solutions.



Figure 2. The location of Open Dumpsite B (a) and the sampling points (b).

Table 1. GPS coordinates of the sampling points				
Sampling Point	Coordinates			
A1	4°30'28"N, 114°01'22"E			
A2	4°30'28"N, 114°01'23"E			
A3	4°30'29"N, 114°01'23"E			
A4	4°30'30"N, 114°01'22"E			
A5	4°30'30"N, 114°01'23"E			
B1	4°30'04"N, 114°00'23"E			
B2	4°30'03"N, 114°00'22"E			
B3	4°30'04"N, 114°00'21"E			
B4	4°30'06"N, 114°00'21"E			
B5	4°30'05"N, 114°00'23"E			

Table 1. GPS coordinates of the sampling points

3. Results and Discussion

3.1. Compositions of waste.

Open dumpsite A is an illegal dumpsite for construction waste (Figure 1). The waste observed at the dumpsite consisted of concrete materials, wood, vegetative waste such as crop residues, stumps, and wood chips, cupboards, sandbags, as well as plastic materials such as polystyrene,

polyvinyl chloride (PVC) pipes, high-density polyethylene (HDPE) pipes, and pipe fittings. Open dumpsite B is an illegal dumpsite for MSW (Figure 2). The waste was composed of plastic materials, batteries, paper waste, metal cans, aluminium cans, nails, screws, metal bars, metal tubes, broken glassware, vegetative waste such as wood chips, leaves, branches, and trunks, broken house furniture, as well as electrical and electronic waste. These open dumpsites were not equipped with proper lining as well as leachate collection and treatment systems. The leachate generated from these dumpsites entered the soil directly. It transported the contaminants, including heavy metals, from the waste into the soil.

3.2. Aluminium concentrations.

As shown in Figure 3, among the first set of samples collected during the initial sampling, the highest level of aluminium recorded was 142.20 mg/kg at A5, while the lowest aluminium concentration recorded was 25.24 mg/kg at location B1. Analyses of the soil samples collected in the second week also showed the highest concentration of aluminium at A5 (135.80 mg/kg) and the lowest concentration of aluminium at B1 (18.66 mg/kg). Except for A2, A3, and A4, the aluminium concentrations at most sampling points decreased in two weeks, with the greatest reduction observed at B3, from 117.20 mg/kg to 81.85 mg/kg. This implied a gradual removal of aluminium from the sampling points, probably due to the leaching and migration of aluminium.



Figure 3. Concentrations of aluminium at different sampling points.

Figure 3 shows that aluminium was detected at all sampling points, though it was more prevalent at the sampling points of the dumpsite for construction waste. The reason could be due to the metallic aluminium and aluminium hydroxide present in concrete [18]. Leaching of aluminium from cookware such as aluminium pots, clay pots, and steel utensils has been reported, and this could account for the presence of aluminium at the MSW dumpsite [19]. The leaching could be facilitated by rainwater which is naturally slightly acidic (pH 4–5.5), as well as acidic soil pH, which could manifest through decomposition of organic materials and imbalanced N, S, and C cycles [20-22]. The aluminium level in A2 increased significantly from 43.56 mg/kg to 61.44 mg/kg, implying a source of aluminium, most likely as a result of continuous release from aluminium-containing waste such as concrete. Nonetheless, these

values are still significantly lower than the typical range of soil aluminium levels (10,000 - 300,000 mg/kg) [20].

3.3. Cadmium concentrations.

As shown in Table 2, the cadmium concentrations at most of the sampling sites were lower than 0.01 mg/kg, except at B2, B3 and B4. B2 recorded the highest initial and Week 2 concentrations of cadmium (0.083 mg/kg and 0.075 mg/kg respectively). Cadmium concentrations were observed to reduce at B2 and B3 over two weeks while marginal increase of cadmium was noted at B4. Elevated cadmium levels were reported at B2 and B3 located at the open dumpsite for MSW.

Location	Mean Concentration (mg/kg)				
	Initial	Week 2			
A1	< 0.01	< 0.01			
A2	< 0.01	< 0.01			
A3	< 0.01	< 0.01			
A4	< 0.01	< 0.01			
A5	< 0.01	< 0.01			
B1	< 0.01	0.019			
B2	0.083 ± 0.014	0.075 ± 0.028			
B3	0.053 ± 0.012	0.049 ± 0.031			
B4	0.019 ± 0.013	0.020 ± 0.018			
B5	< 0.01	< 0.01			

Table 2. Cadmium (Cd) concentrations in the soil samples collected from different sampling points.

MSW is generally more complex in composition than construction waste, consisting of food waste, household waste, garden waste, textiles, diapers, and even hazardous waste such as electrical and electronic waste [23]. Batteries were sighted at the dumpsite, and nickel-cadmium batteries with cadmium as the anode might contribute to the cadmium content in soil once leached. However, cadmium is also naturally occurring in soil, with concentrations up to 5 g/L in soil water as a result of weathering [24]. In fact, Cd is usually present in soil at 0.01 to 1 mg/kg and the global mean of its concentration in soil is 0.36 mg/kg [25]. Therefore, while a few sampling points showed elevated Cd levels, they are considered to fall within the common concentration range of Cd in soil.

3.4. Copper concentrations.

Figure 4 shows that B3 contained the highest amounts of copper both during initial sampling (14.99 mg/kg) and sampling on Week 2 (13.78 mg/kg), but there was a slight decrease in copper over two weeks. Both soil samples taken from B2 showed that the samples contained the second highest amounts of copper (5.01 mg/kg and 3.47 mg/kg). A larger decline in copper concentration was observed at B2 (1.54 mg/kg) in the second week. Other sampling points had relatively low copper concentrations. Two sampling points with high copper concentrations were located at the dumpsite for MSW (Figure 4). While the high copper concentrations could be linked to the composition of MSW, the distribution of copper in soil is multifactorial, with climate and geology all playing a role [26]. Agricultural activities could also introduce copper into the soil. A study examining the soil copper concentrations in the European Union found the mean copper concentration to be 16.85 mg/kg and vineyards were generally associated with higher mean soil copper concentrations [27]. In most of the sampling points, copper

concentrations decreased with time. As a plant micronutrient, copper could be taken up by plants [28]. A decrease in soil copper concentration could also be caused by high soil organic carbon and high clay content [28]. Nonetheless, copper is not as mobile as aluminium and would only be subjected to leaching in extremely sandy soils [29].



Figure 4. Concentrations of copper at different sampling points.

3.5. Iron concentrations.

At B2 and B3, there were markedly higher initial iron concentrations than other sampling points, with B3 recording the highest initial iron concentration of 241.77 mg/kg, followed by B2 at 233.90 mg/kg (Figure 5). The iron concentration from the first sampling practice at A1 came in third with 108.33 mg/kg and that at A5 came in fourth with 99.03 mg/kg (Figure 5). The prevalence of iron concentrations as revealed from the second sampling in Week 2 was similar where B3 had the highest iron concentration (241.67 mg/kg), followed by B2 (238.43 mg/kg). However, a significant decrease in the iron level at A1 to 82.93 mg/kg pushed it to the fourth place after A5 (93.50 mg/kg).



Figure 5. Concentrations of iron at different sampling points.

Six sampling points showed reduced iron concentrations while only four sampling points showed increased iron concentrations over two weeks. Like aluminium, iron was detected at all sampling points at relatively high concentrations, and the highest soil iron concentrations were reported at B2 and B3, similar to copper. A plausible reason for the prevalence of iron at the sampling points is that iron is commonly found in construction and municipal waste [30]. Besides, MSW contains a significant proportion of metals, and iron is one of the most common metals reported [23]. The changes in soil iron levels at the sampling points over two weeks were highly variable, with A1 showing an obvious reduction. The changes could be attributed to factors such as soil organic matter and pH. Iron is also a plant micronutrient, and the bioaccumulation of iron by plants is likely to vary with the species of plants [31].

3.6. Manganese concentrations.

Referring to Figure 6, most of the sampling points had low manganese concentrations, except B2 and B3. Analyses of the first set of soil samples revealed that B2 and B3 had manganese concentrations of 9.62 mg/kg and 6.24 mg/kg, respectively. The manganese concentrations at other sampling points were below 1.1 mg/kg (Figure 6). Over two weeks, B2 saw a more-than-twofold increase in manganese concentration to 22.60 mg/kg, while the manganese concentration at B3 decreased slightly (Figure 6).



Figure 6. Concentrations of manganese at different sampling points.

Manganese is naturally present in soil at concentrations of 20 to 3000 ppm, or 20–3000 mg per kg of soil. It is usually present as Mn^{2+} in soil with the tendency to form complexes with organic compounds [32]. Its availability in soil is affected by pH, and lower pH results in higher solubility, hence availability to plants [32]. This study again points to significantly higher manganese levels at the MSW dumpsite, probably due to the more diverse composition of MSW. Electrical and electronic waste, which was also disposed of at the dumpsite, might have been a source of manganese. The proximity of the waste to B2 and B3 has possibly led to the elevated soil manganese. Nonetheless, at the highest recorded level of 22.60 mg/kg, the soil manganese concentration is within the natural range [33]. The significant increase in soil

manganese at B2 over two weeks might imply a source of manganese, most likely from the electrical and electronic waste there.

3.7. Nickel concentrations.

Among the first set of soil samples, the nickel concentration at B3 (1.77 mg/kg) was the highest, followed by B2 (1.14 mg/kg), whereas that at B1 was the lowest (0.02 mg/kg) (Figure 7). In Week 2, the nickel concentration at B3 remained the highest (1.73 mg/kg). However, the nickel concentration at B2 had decreased substantially to 0.41 mg/kg (Figure 7). B2 and B3 were located at the open dumpsite receiving MSW. Nickel has been detected in soil at concentrations between 4 and 80 ppm [34]. Nickel emissions into the environment have been linked to nickel refining, steel production, coal combustion, and municipal incineration [4]. In regards to the dumpsite receiving MSW. For instance, rechargeable nickel-cadmium batteries are a potential source of both nickel and cadmium [35]. It is interesting to note the considerable decrease in the nickel level at B2 over two weeks. In fact, nickel has relatively low mobility, equivalent to chromium and zinc in calcareous loam due to its tendency to sorb into the solid phase of soil [34]. The reduction could be a result of the sorption of nickel causing its uneven distribution at B2.



Figure 7. Concentrations of nickel at different sampling points.

3.8. Zinc concentrations.

As with copper, manganese and nickel, B2 and B3 recorded significantly higher amounts of zinc in the first set (35.03 mg/kg and 21.17 mg/kg, respectively) and the second set (29.83 mg/kg and 20.47 mg/kg, respectively) of soil samples (Figure 8). The zinc concentrations at other sampling points were less than 0.80 mg/kg (Figure 8). B2 showed a notable reduction of zinc levels over two weeks, similar to that observed for nickel, except that the extent of reduction for zinc was far less. Zinc has soil mobility that is similar or closely resembles that of nickel, hence a tendency to adsorb to soil particles [34]. The reduction of zinc at B2 could also be a consequence of the uneven distribution of zinc, in addition to other factors mentioned

earlier, such as soil types, organic content, and soil pH, that influence the distribution and availability of heavy metals. The typical mean soil zinc level is 50 ppm, so the levels of zinc reported are not alarming [36]. Zinc is also a micronutrient needed by plants. Zinc is a common component of electrical and electronic waste, which is likely the reason that it was detected at higher concentrations at B2 and B3 with such waste in their vicinity [36]. It is noteworthy that the incineration of MSW has been reported to produce bottom ash containing significant amounts of copper, lead, and zinc. This implies the common presence of copper and zinc in MSW [37].



Figure 8. Concentrations of zinc at different sampling points.

3.9. Comparison with the soil screening levels.

There is currently a lack of regulation on the acceptable levels of heavy metals in soil, developed for Malaysia. However, the Department of Environment Malaysia published the 'Contaminated Land Management and Control Guidelines No. 1: Malaysian Recommended Site Screening Levels for Contaminated Land', which contains the Soil Screening Levels (SSLs) adopted from the US EPA Regional Screening Levels [38]. These SSLs are used to determine if the levels of heavy metals at the sampling points are of concern. According to Table 3, the highest soil levels of the respective heavy metals were well below the SSLs, showing that the levels do not constitute a concern for soil contamination. Therefore, the sampling points are currently not considered contaminated sites.

Heavy Metal	Highest Level Reported (mg/kg)	SSL for Residential Soil* (mg/kg)
Aluminium	142.20 ± 0.13	77000
Cadmium	0.083 ± 0.014	7
Copper	14.99 ± 0.01	3100
Iron	241.77 ± 0.02	55000
Manganese	22.60 ± 0.01	1800
Nickel	1.77 ± 0.001	1600
Zinc	35.03 ± 0.002	23000

Table 3. The highest levels of heavy metals reported and their corresponding SSLs.

*SSLs for residential soil are used because the dumpsites are located at the vicinity of residential areas or a closer proximity to residential areas as shown in Figures 1 and 2.

Having said that, it is crucial to regulate illegal dumping activities due to the health and safety concerns they pose, particularly in breeding vermin and pests, as well as the physical risks related to slipping, tripping, puncturing, etc. [39, 40]. Besides, illegal dumpsites pose significant aesthetic impacts and release odors, which could cause discomfort to the receptors nearby [41]. The presence of these dumpsites also encourages irresponsible waste disposal, and it is difficult to control the types of waste received by these dumpsites.

3.10. Comparison with other dumpsites and correlation analyses.

Table 4 shows that the levels of soil heavy metals reported in this study are generally lower compared to other studies, with the Cu, Fe, and Mn levels at the lower ends of those reported by Syed Ismail et al. for four non-sanitary landfills located in the Langat Water Catchment Malaysia [42]. The concentrations of Al and Cd were significantly lower than those in the soils of the four landfills, while the concentration of Ni and Zn were comparable to those of the landfills, with the highest Zn concentration exceeding that of the landfills. The levels of soil Al and Zn in the dumpsites studied were close to the lower ends of those reported for a municipal landfill at Simpang Renggam, Johor. The Ni concentrations, however, were significantly lower than those of the municipal landfill.

Heavy Metal	Range Reported in This Study (mg/kg)	Range for Non-Sanitary Landfills [42] (mg/kg)	Range for a Municipal Landfill [43] (mg/kg)		
Aluminium	24.67 - 142.20	389 - 1,450	Not tested		
Cadmium	< 0.01 - 0.083	2.57 - 4.61	Not tested		
Copper	0.10 - 14.99	13.7 - 43.2	14 - 74		
Iron	11.20 - 241.77	299 - 1,260	Not tested		
Manganese	0.09 - 22.60	19.7 - 90.8	Not tested		
Nickel	0.02 - 1.77	0.28 - 1.55	16 - 35		
Zinc	0.14 - 35.03	6.47 - 17.8	32 - 78		

Table 4. Comparison of the levels of soil heavy metals with those reported in other studies

Pearson's correlations in Table 5 confirm the findings that the initial levels of Cd, Cu, Fe, Mn, Ni and Zn were significantly interrelated, and this is shown in Figures 4-8 and Table 2 that consistently high initial levels of soil Cd, Cu, Fe, Mn, Ni and Zn were detected at B2 and B3. No significant correlations were observed between Al and other heavy metals likely because of the obvious differences between the pattern of Al distribution and those of other heavy metals across the sampling points.

Table 5. Pearson's correlations for the initial concentrations of soil heavy metals.

						5	
Heavy Metal	Initial Al	Initial Cd	Initial Cu	Initial Fe	Initial Mn	Initial Ni	Initial Zn
Initial Al	1	0.209	0.408	0.532	0.264	0.413	0.209
Initial Cd	-	1	0.689*	0.861**	0.986**	0.849**	0.993**
Initial Cu	-	-	1	0.809**	0.716*	0.962**	0.704*
Initial Fe	-	-	-	1	0.910**	0.916**	0.880**
Initial Mn	-	-	-	-	1	0.876**	0.996**
Initial Ni	-	-	-	-	-	1	0.865**
Initial Zn	-	-	-	-	-	-	1
Initial Zn	-	-	-	-	-	-	1

**Significant at p = 0.01; *significant at p = 0.05

A similar interrelation between the concentrations of Cd, Fe, Mn and Zn was observed except that the correlations between Cd and Cu, as well as between Cd and Ni became insignificant, particularly the latter, likely due to the changes in the concentrations of Cu, and Ni especially, by Week 2 (Table 6). Similarly, the substantial increase in soil Mn concentration (Figure 6) and the substantial decrease in soil Ni concentration in Week 2 (Figure 7) rendered the Mn-Ni correlation insignificant. The change in soil Mn level also caused the Mn-Cu correlation to become insignificant.

						5	
Heavy Metal	Wk2 Al	Wk2 Cd	Wk2 Cu	Wk2 Fe	Wk2 Mn	Wk2 Ni	Wk2 Zn
Wk2 Al	1	-0.028	0.160	0.244	-0.088	0.237	-0.009
Wk2 Cd	-	1	0.630	0.893**	0.928**	0.596	0.987**
Wk2 Cu	-	-	1	0.786**	0.335	0.996**	0.690*
Wk2 Fe	-	-	-	1	0.774*	0.786**	0.930**
Wk2 Mn	-	-	-	-	1	0.300	0.913**
Wk2 Ni	-	-	-	-	-	1	0.660*
Wk2 Zn	-	-	-	-	-	-	1

Table 6. Pearson's correlations for Week 2 concentrations of soil heavy metals.

**Significant at p = 0.01; *significant at p = 0.05

3.11. Limitations.

This study has its limitations. While endeavoring to include as many heavy metals as possible in the study, some commonly reported heavy metals such as lead, mercury, and chromium have been excluded due to technical constraints. However, the number of heavy metals analyzed is comparable to that of other studies [4, 44–46]. The sampling points were selected to ensure that they were sufficiently representative of the dumpsites. Even so, it is likely that certain crucial points have not been captured [47]. This study primarily aims to examine the levels of heavy metals in the soil of illegal dumpsites and is therefore limited in describing the temporal changes of these levels. Only two sets of samples were collected on a two-week interval from the sampling points. This study did not extend the variation tracking of the heavy metals beyond that, nor did it perform the tracking on a shorter interval. On the same note, this study did not perform a detailed inventory of the types of waste spotted at each sampling point and the quantities.

4. Conclusions

This study elucidates the levels of heavy metals in the soil in and around two illegal open dumpsites, one receiving construction waste and the other receiving MSW. It shows that aluminium, copper, iron, manganese, nickel, and zinc have been detected at all the sampling points, whereas cadmium at some sampling points had levels below the detection limit. The levels of each heavy metal vary at each sampling point, with two sampling points at the dumpsite receiving MSW demonstrating elevated levels of cadmium, copper, iron, manganese, nickel, and zinc. This was seemingly associated with the compositional complexity of MSW, especially the presence of electrical and electronic waste, including batteries, which contained an array of heavy metals. The prevalence of aluminium is similar in both the dumpsites, indicating that construction waste like concrete might contain leachable aluminium components. This study shows that the current levels of heavy metals tested were not of significant concern and are not likely to constitute soil contamination. Nonetheless, the dumpsites could become breeding grounds for vermin and pose aesthetic impacts. It is advisable that illegal dumping in the region should be controlled with existing illegal dumpsites cleared. Since the types of waste sent to illegal dumpsites are not regulated, there is always a risk of site contamination.

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Competing Interest

The authors declare that there is no competing interest.

References

- Tang, K.H.D. (2021). Phytoextraction of Lead: Its Feasibility, Constraints and Concerns. Asian Soil Research Journal, 5(4), 1–9. <u>https://doi.org/10.9734/asrj/2021/v5i430113</u>.
- [2] Duffus, J.H. (2002). Heavy metals'a meaningless term. *Pure and Applied Chemistry*, 74(5), 793-807. <u>http://dx.doi.org/10.1351/pac200274050793</u>.
- [3] Tchounwou, P.B.; Yedjou, C.G.; Patlolla, A.K.; Sutton, D.J. (2012). Heavy metals toxicity and the environment. *Experientia Supplementum*, 101, 133–164. <u>https://doi.org/10.1007/978-3-7643-8340-4</u>.
- [4] Ngole, V.M.; Ekosse, G.I.E. (2012). Copper, nickel and zinc contamination in soils within the precincts of mining and landfilling environments. *International Journal of Environmental Science and Technology*, *9*(3), 485–494. <u>https://doi.org/10.1007/s13762-012-0055-5</u>.
- [5] Tang, Y.Y., Tang, K.H.D.; Maharjan, A.K.; Aziz, A.A.; Bunrith, S. (2021). Malaysia moving towards a sustainability municipal waste management. *Industrial and Domestic Waste Management*, 1(1), 26–40. <u>http://doi.org/10.53623/idwm.v1i1.51</u>.
- [6] Alzamora, B.R.; de V. Barros, R.T. (2020). Review of municipal waste management charging methods in different countries. *Waste Management*, 115, 47–55. <u>https://doi.org/10.1016/j.wasman.2020.07.020</u>.
- [7] Waste to energy for a sustainable future. (accessed on 1 Augusts 2022) Available online: https://www.mida.gov.my/waste-to-energy-for-a-sustainable-future/.
- [8] Zainun, N.Y.; Rahman, I.A.; Rothman, R.A. (2016). Mapping Of Construction Waste Illegal Dumping Using Geographical Information System (GIS). *IOP Conference Series: Materials Science and Engineering*, *160*, 12049. <u>https://doi.org/10.1088/1757-899x/160/1/012049</u>.
- [9] Ismail, Z.; Abdullah, S.Z.; Othman, S.Z.; Shirazi, S.M.; Karim, R. (2013). Assessment of the Relative Adequacy of Landfills as a Means of Solid Waste Disposal in Malaysia. *CLEAN – Soil, Air, Water, 41*(11), 1122–1128. <u>https://doi.org/https://doi.org/10.1002/clen.201200316</u>.
- [10] Hussein, M.; Yoneda, K.; Mohd-Zaki, Z.; Amir, A.; Othman, N. (2021). Heavy metals in leachate, impacted soils and natural soils of different landfills in Malaysia: An alarming threat. *Chemosphere*, 267, 128874. <u>https://doi.org/10.1016/j.chemosphere.2020.128874</u>.
- [11] Agamuthu, P.; Fauziah, S.H. (2010). Heavy metal pollution in landfill environment: A Malaysian case study. 2010 4th International Conference on Bioinformatics and Biomedical Engineering, 1–4. <u>https://doi.org/10.1109/ICBBE.2010.5516886</u>.
- [12] Adama, M.; Esena, R.; Fosu-Mensah, B.; Yirenya-Tawiah, D. (2016). Heavy Metal Contamination of Soils around a Hospital Waste Incinerator Bottom Ash Dumps Site. *Journal of Environmental and Public Health*, 2016, 8926453. <u>https://doi.org/10.1155/2016/8926453</u>.
- [13] Peng, N.; Li, Y.; Liu, T.; Lang, Q.; Gai, C.; Liu, Z. (2017). Polycyclic Aromatic Hydrocarbons and Toxic Heavy Metals in Municipal Solid Waste and Corresponding Hydrochars. *Energy & Fuels*, 31(2), 1665–1671. <u>https://doi.org/10.1021/acs.energyfuels.6b02964</u>.
- [14] Cittadino, A.; Ocello, N.; Majul, M.V.; Ajhuacho, R.; Dietrich, P.; Igarzabal, M.A. (2020). Heavy metal pollution and health risk assessment of soils from open dumps in the Metropolitan Area of

Buenos Aires, Argentina. *Environmental Monitoring and Assessment*, 192(5), 291. https://doi.org/10.1007/s10661-020-8246-x.

- [15] Ogunbanjo, O.; Onawumi, O.; Gbadamosi, M.; Ogunlana, A.; Anselm, O. (2016). Chemical speciation of some heavy metals and human health risk assessment in soil around two municipal dumpsites in Sagamu, Ogun state, Nigeria. *Chemical Speciation & Bioavailability*, 28(1–4), 142– 151. https://doi.org/10.1080/09542299.2016.1203267.
- [16] Agbeshie, A.A.; Adjei, R.; Anokye, J.; Banunle, A. (2020). Municipal waste dumpsite: Impact on soil properties and heavy metal concentrations, Sunyani, Ghana. *Scientific African*, 8, e00390. <u>https://doi.org/10.1016/j.sciaf.2020.e00390</u>.
- [17] Wijesekara, S.S.R.M.D.H.R.; Mayakaduwa, S.S.; Siriwardana, A.R.; de Silva, N.; Basnayake, B.F.A.; Kawamoto, K.; Vithanage, M. (2014). Fate and transport of pollutants through a municipal solid waste landfill leachate in Sri Lanka. *Environmental Earth Sciences*, 72(5), 1707–1719. https://doi.org/10.1007/s12665-014-3075-2.
- [18] Mary Joseph, A.; Snellings, R.; Nielsen, P.; Matthys, S.; De Belie, N. (2020). Pre-treatment and utilisation of municipal solid waste incineration bottom ashes towards a circular economy. *Construction and Building Materials*, 260, 120485. <u>https://doi.org/https://doi.org/10.1016/j.conbuildmat.2020.120485</u>.
- [19] Odularu, A.T.; Ajibade, P.A.; Onianwa, P.C. (2013). Comparative Study of Leaching of Aluminium from Aluminium, Clay, Stainless Steel, and Steel Cooking Pots. *ISRN Public Health*, 2013, 517601. <u>https://doi.org/10.1155/2013/517601</u>.
- [20] Bojórquez-Quintal, E.; Escalante-Magaña, C.; Echevarría-Machado, I.; Martínez-Estévez, M. (2017). Aluminum, a Friend or Foe of Higher Plants in Acid Soils. *Frontiers in Plant Science*, 8, 1-18. <u>https://doi.org/10.3389/fpls.2017.01767</u>.
- [21] Tang, K.H.D.; Kristanti, R.A. (2022). Bioremediation of perfluorochemicals: current state and the way forward. *Bioprocess and Biosystems Engineering*. <u>https://doi.org/10.1007/s00449-022-02694-z</u>.
- [22] Li, C.; Tang, K.H.D. (2023). Effects of pH and Temperature on the Leaching of Di (2-Ethylhexyl) Phthalate and Di-n-butyl Phthalate from Microplastics in Simulated Marine Environment. *Biointerface Research in Applied Chemistry*, 13(3). <u>https://doi.org/10.33263/BRIAC133.269</u>.
- [23] Tang, K.H.D. (2020). Municipal Solid Waste Management in the Sarawak State of Malaysia and the Way Forward. Asian Journal of Environment & Ecology, 12(2), 38–55.
- [24] Smolders, E.; Brans, K.; Foldi, A.; Merckx, R. (1999). Cadmium fixation in soils measured by isotopic dilution. Soil Science Society of American Journal, 63. <u>https://doi.org/10.2136/sssaj1999.03615995006300010013x</u>.
- [25] Kubier, A.; Wilkin, R.T.; Pichler, T. (2019). Cadmium in soils and groundwater: A review. Applied Geochemistry, 108, 104388. <u>https://doi.org/10.1016/j.apgeochem.2019.104388</u>.
- [26] Tang, K.H.D., Awa, S.H.; Hadibarata, T. (2020). Phytoremediation of Copper-Contaminated Water with Pistia stratiotes in Surface and Distilled Water. *Water, Air, & Soil Pollution, 231*(12), 573. <u>https://doi.org/10.1007/s11270-020-04937-9</u>.
- [27] Ballabio, C.; Panagos, P.; Lugato, E.; Huang, J.H.; Orgiazzi, A.; Jones, A.; Fernández-Ugalde, O.; Borrelli, P.; Montanarella, L. (2018). Copper distribution in European topsoils: An assessment based on LUCAS soil survey. *Science of The Total Environment, 636*, 282–298. <u>https://doi.org/10.1016/j.scitotenv.2018.04.268</u>.
- [28] Lange, B.; van der Ent, A.; Baker, A.J.M.; Echevarria, G.; Mahy, G.; Malaisse, F.; Meerts, P.; Pourret, O.; Verbruggen, N.; Faucon, M.P. (2017). Copper and cobalt accumulation in plants: a critical assessment of the current state of knowledge. *New Phytologist*, 213(2), 537–551. <u>https://doi.org/10.1111/nph.14175</u>.

- [29] Sun, Q.; Li, T.; Alva, A.K.; Li, Y.C. (2019). Mobility and fractionation of copper in sandy soils. *Environmental Pollutants and Bioavailability*, 31(1), 18–23. <u>https://doi.org/10.1080/09542299.2018.1558114</u>.
- [30] Li, W.B.; Yao, J.; Tao, P.P.; Guo, M.T.; Feng, X.Y.; He, Y.N.; Fang, C.R.; Shen, D.S. (2010). A comparative study on two extraction procedures in speciation of iron in municipal solid waste. *Journal of Hazardous Materials*, 182(1), 640–648. <u>https://doi.org/10.1016/j.jhazmat.2010.06.080</u>.
- [31] Guerinot, M.L.; Yi, Y. (1994). Iron: Nutritious, Noxious, and Not Readily Available. Plant Physiology, 104(3), 815–820. <u>https://doi.org/10.1104/pp.104.3.815</u>.
- [32] Sarkar, D.; De, D.K.; Das, R.; Mandal, B. (2014). Removal of organic matter and oxides of iron and manganese from soil influences boron adsorption in soil. *Geoderma*, 214–215, 213–216. <u>https://doi.org/10.1016/j.geoderma.2013.09.009</u>.
- [33] Rai, R.K.; Singh, V.P.; Upadhyay, A. (2017). Chapter 2 Irrigation Project Planning. In Planning and Evaluation of Irrigation projects; Rai, R.K., Singh, V.P., Upadhyay, A., Eds.; Academic Press: Cambridge, USA, pp. 7–24. <u>https://doi.org/10.1016/B978-0-12-811748-4.00002-9</u>.
- [34] Holmgren, G.G.S.; Meyer, M.W.; Chaney, R.L.; Daniels, R.B. (1993). Cadmium, lead, zinc, copper, and nickel in agricultural soils of the United States of America. *Journal of Environmental Quality*, 22, 335-348. <u>https://doi.org/10.2134/jeq1993.00472425002200020015x</u>.
- [35] Sorahan, T.; Esmen, N.A. (2004). Lung cancer mortality in UK nickel-cadmium battery workers, 1947–2000. Occupational and Environmental Medicine, 61(2), 108 LP – 116. <u>https://doi.org/10.1136/oem.2003.009282</u>.
- [36] Barrow, N.J. (1993). Mechanisms of Reaction of Zinc with Soil and Soil Components BT Zinc in Soils and Plants. In Proceedings of the International Symposium on Zinc in Soils and Plants, The University of Western Australia, Australia, 27–28 September 1993; Robson, A.D., Ed.; Springer: Amsterdam, Netherlands, pp. 15–31).. <u>https://doi.org/10.1007/978-94-011-0878-2_2</u>.
- [37] Seniunaite, J.; Vasarevicius, S. (2017). Leaching of Copper, Lead and Zinc from Municipal Solid Waste Incineration Bottom Ash. *Energy Procedia*, 113, 442–449. <u>https://doi.org/10.1016/j.egypro.2017.04.036</u>.
- [38] Contaminated Land Management and Control Guidelines No.1: Malaysian Recommended Site Screening Levels for Contaminated Land. (accessed on 1 Augusts 2022) Available online: <u>https://www.doe.gov.my/wp-content/uploads/2021/07/Contaminated-Land-Management-and-Control-Guidelines-No-1_Malaysian-Recommended-Site-Screening-Levels-for-Contaminated-Land.pdf</u>.
- [39] Tang, K.H.D. (2019). The Dilemma of Environmental Impact Assessment in Sarawak, Malaysia. *Journal of Sustainability Science and Management*, *14*(3), 3–6.
- [40] Tang, K.H.D. (2020). A comparative overview of the primary Southeast Asian safety and health laws. *International Journal of Workplace Health Management*, 13, 601-632. <u>https://doi.org/10.1108/IJWHM-10-2019-0132</u>.
- [41] Tang, K.H.D. (2020). A Case Study of The Environmental Impact Assessment Legislations In Sarawak, Malaysia. Asia Pacific *Journal of Energy and Environment*, 7, 47–54. <u>https://doi.org/10.18034/apjee.v7i1.273</u>.
- [42] Syed Ismail, S.N.; Ishak, C.F.; Samah, M.A.A.; Hatta, E.M.; Wahab, A.S.A. (2015). Soil contamination from non-sanitary waste landfill in Langat water catchment area, Malaysia. *Journal* of Scientific Research and Reports, 7(6), 480–493.
- [43] Zaidi, E.; FahrulRazi, M.J.; Azhar, A.T.S.; Hazreek, Z.A.M.; Shakila, A.; Norshuhaila, M.S.; Omeje, M. (2017). Radionuclides (40K,232Th and238U) and Heavy Metals (Cr, Ni, Cu, Zn, As and Pb) Distribution Assessment at Renggam Landfill, Simpang Renggam, Johor, Malaysia. IOP Conference Series: Materials Science and Engineering, 226, 12070. <u>https://doi.org/10.1088/1757-899x/226/1/012070</u>.

- [44] Chen, W.; Li, L.; Chang, A.C.; Wu, L.; Chaney, R.L.; Smith, R. (2009). Characterizing the solidsolution partitioning coefficient and plant uptake factor of As, Cd, and Pb in California croplands. Agriculture, *Ecosystems and Environment*, 129, 212-220. https://doi.org/10.1016/j.agee.2008.09.001.
- [45] Grosbois, C.; Meybeck, A.; Horowitz, A.; Ficht, A. (2006). The spatial and temporal trends of Cd, Cu, Hg, Pb and Zn in Seine River floodplain deposits (1994–2000). *Science of the Total Environment*, 356. <u>https://doi.org/10.1016/j.scitotenv.2005.01.049</u>.
- [46] Buekers, J.; Degryse, F.; Maes, A.; Smolders, E. (2008). Modelling the effects of ageing on Cd, Zn, Ni and Cu solubility in soils using an assemblage model. *European Journal of Soil Science*, 59, 22-37. <u>https://doi.org/10.1111/j.1365-2389.2008.01053.x</u>.
- [47] Tang, K.H.D. (2019). Phytoremediation of Soil Contaminated with Petroleum Hydrocarbons: A Review of Recent Literature. *Global Journal of Civil and Environmental Engineering*, 1, 33–42. <u>https://doi.org/10.36811/gjcee.2019.110006</u>.



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