STUDY ON HEAVY METAL CONTAMINATION DISTRIBUTION AT ACTIVE LANDFILL AT DIFFERENT DEPTHS AND RADIUSES

OTHMAN, R.[1*] – MOHD LATIFF, N. H.[1] – BAHARUDDIN, Z. M.[1] – HASHIM, K. S. H. Y.[2] – LUKMAN HAKIM MAHAMOD, L. H.[3]

[1]*International Institute for Halal Research and Training (INHART)*
Department of Landscape Architecture, Kulliyyah of Architecture and Environmental Design (KAED), International Islamic University Malaysia, 53100 Kuala Lumpur, Malaysia

[2]*Department of Urban & Regional Planning, Kulliyyah of Architecture and Environmental Design (KAED), International Islamic University Malaysia, 53100 Kuala Lumpur, Malaysia*

[3]*Department of Quantity Surveying, Kulliyyah of Architecture and Environmental Design (KAED), International Islamic University Malaysia, 53100 Kuala Lumpur, Malaysia*

[*]*Corresponding author*
e-mail: rashidi@iium.edu.my; phone: +60-126-644-772; fax: +60-361-964-864

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Abstract. Landfilling is more preferable in Malaysia compared to another disposal method due to low cost, and availability of land. Other than solid waste, the percolation of water into the landfill leads to leachate formation. The migration of waste in leachate form may accelerate the heavy metal contamination of the soil one of the major concerns in landfilling. This study aimed in comparing soil samples taken from five different sites in Selangor of inert waste (Sungai Kertas, Kuang and Dengkil) and sanitary (Tanjung Dua Belas and Jeram) landfills at different depths (0-30 cm, 30-60 cm and 60-90 cm) and radiuses (5-10 m, 10-15 m and 15-20 m), for ten heavy metals (Al, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd and Pb) to find the risk of heavy metal movement from the upper layer cell into the deeper layer of the soil block. All the data were analysed using ICP-MS (Perkin Elmer NexION 300X). Al and Fe displayed high concentration at most of the sites especially at the deeper depth of the soil.
Keywords: *urban pollution, landfill, active sanitary landfills, inorganic pollutant, leachate, municipal solid waste*

Introduction

According to Agamuthu et al. (2014) and Yusoff et al. (2013), there is a total of about 251 landfills of different sizes and ages recognized officially in Malaysia. Out of these, 111 were closed and 150 are active landfills. These active landfills include 77 open dumpsites, 49 open-tipping sites, 19 landfills without leachate treatment, 10 landfills with leachate treatment, and 6 are sanitary (engineered). In Malaysia, proper disposal of solid waste at sanitary landfills constitute only a small portion of a whole and was built recently compared to open dumps which constitute around 90% of all the waste disposal sites (Agamuthu and Fauziah, 2011; Sharifah and Latifah, 2013).

Selangor as one of the most developed states in Malaysia has produced the highest MSW per capita due to the high standard of living (Agamuthu, 2001; Leete, 2002). Studies reported that Selangor is responsible for one-third of the total amount of solid waste generated in Malaysia with 3,923 tons daily (Riber et al., 2005; Ismail and Manaf, 2013; Sharifah and Latifah, 2013). Twelve landfill sites from a total of 20 landfill sites in Selangor (closed and operating) are considered as open dumpsites with level 0 sanitary

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classification, which generate the highest risk of contamination and leachate migration. In addition, all 20 landfills are located in 5 main river basins where the Sungai Selangor Basin has the highest number of operating landfills (three) and four closed landfills. The classification is as followings; Level 0: Open dumping; Level I: Controlled tipping; Level II: Sanitary landfill with a bund and daily soil cover; Level III: Sanitary landfill with leachate recirculation system; and Level IV: Sanitary landfill with leachate treatment facilities.

Sanitary landfilling and open dumping are commonly known across the world as disposal of municipal solid waste (MSW) method. Sanitary landfilling is predominantly practiced in developed nations while open dumping is common in low-income and developing nations (Yahaya, 2016). An open dump is a primitive and non-sanitary waste disposal site where indiscriminate wastes are improperly tipped or dumped in a pre-existing hole or on the side of a hill without proper maintenance (Agamuthu, 2001; Blight, 2008). As an alternative, a sanitary landfill utilizes the principles of engineering to ensure the protection of the environment and public health while providing safe long-term disposal of solid wastes (American Society of Civil engineers, 1959). In Malaysia, municipal solid waste is disposed of in both the newer sanitary landfills and older open dumps.

The increasing of waste generation due to this municipal solid waste, however, shortens the span of a landfill. Thus, more and more new areas have to be converted into disposal sites. The negative impacts of MSW landfill on the environment cause a wide range of concern. It includes the risk of explosion, odor problem, leachate seeping into surface and groundwater system, as well as, soil contamination due to heavy metal sourced from disposed waste (Fauziah and Agamuthu, 2005). Contaminated soil remediation is reported as one of the most expensive technologies in environmental management.

Based on the previous study, most organic chemical substances will eventually either be degraded through biochemical reactions in the landfill or be leached out of the landfill with water movement. However, the majority of heavy metals will remain in the landfill because heavy metal migration is very limited compared to the number of metals accumulated in the landfill (Øygard et al., 2004; Riber et al., 2004), especially in anaerobic processes. The slow movement of heavy metals is the result of heavy metals being subjected to strong sorption on soil particles, precipitation under anaerobic conditions, and chelation with inorganic and organic ligands in landfills (Bozkurt et al., 1999, 2000). Heavy metal contamination is mainly due to the subsequent migration of leachate from and within the landfill's waste cell. The natural process taking place within the boundaries of the waste cells accelerate the process of heavy metal leaching from the waste component. Various types of wastes are the main sources of heavy metal within a landfill system. This includes metal waste components such as food cans and scrap metal and the indiscriminate dumping of household hazardous waste and electronic waste such as batteries and old computer. The release of heavy metal into the adjacent environment is a serious environmental concern and a threat to public health and safety.

Material and methods

Study area description

Five active landfill sites (3 non-sanitary landfills and 2 sanitary landfills) located at Selangor, Malaysia namely; Sungai Kertas non-sanitary landfill (3°15'52.5"N

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101°41'05.1"E), Kuang non-sanitary landfill (3°15'31.4"N 101°33'50.0"E), Dengkil non-sanitary landfill (2°52'12.2"N 101°38'54.1"E), Tanjung Dua Belas sanitary landfill (2°43'48.9"N 101°36'21.7"E) and Jeram sanitary landfill (3°11'23.8"N 101°21'54.3"E) were selected for this study. The site selection was based on the types of the landfill and the size of the landfill located in Selangor state. *Table 1* indicates the landfill sites info with the area covered, wasted collected, total year of operation and the current status of the landfill studied.

Table 1. *Selected landfill sites with the area covered, waste collected per day, total year operation and current status*

Landfill Sites	Area (Acre)	Waste Collected (Tone/Day)	Total Year Operation	Current Status	Source
Kampung Sungai Kertas Inert Waste	14.19	130	7	Active	Othman et al. (2016)
Kuang Inert Waste Landfill	27	50	11	Active	Tengku Ibrahim et al. (2017)
Dengkil Inert Waste Landfill	145	400	25	Active	Othman et al. (2016)
Tanjung 12 Sanitary Landfill	160	1000	20	Active	Yahaya et al. (2016)
Jeram Sanitary Landfill	160	2500	16	Active	Othman et al. (2016)

Soil sampling

Soil samples were collected at different points (0-200 mm depth, approx. 1000 g) were taken by using soil auger (Eijkelkamp Agrisearch). The soil then was sealed in a polyethylene bag and labeled. The soil was dried in an oven for 70ºC for 3 days to a week depending on the moistness of the soil. Then samples were ground by using agate mortar until becoming small particles. Then it was sieved using a 2 mm mesh to remove stones and plant materials. Then samples were stored at room temperature before being digested using Microwave Digestion Ethos D (Milestone, 2001). Heavy metals, Al, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb were then analysed by using ICP-MS (Perkin Elmer NexION 300X).

Heavy metals digestion and analysis

0.5 g of soil samples were accurately weighed into a container made of PFA perfluoroalkoxy polymer and digested through a microwave digestion system using the digestion method as described by Zhao et al. (1994). Soil samples were air-dried and sieved. Soil samples were passed through a 2 mm sieve and ready for further analysis. Dried and the ground sample was mixed with 10 ml of concentrated nitric acid (HNO_3 65%) and digested. Acid was added for each soil samples and then the digestion tubes were placed in a rotor segment by using a torque wrench. The segments were inserted into the microwave cavity and connected with the temperature sensor. The mixture temperature was adjusted to ±175ºC and 1,200 Watt of power for 30 minutes using Microwave Digestion (Milestone Start D) as detailed in Method US EPA 3051. The digestion was completed after the last solution was clear and no brownish fumes were released from the digestion vessel tubes. When digestion was completed, samples

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were removed and diluted. The soil digests were adjusted to the final volume of 50 mL with deionized water. This solution is further 1:1 diluted for the analysis of components by ICP-MS and divided into triplicate each into 15 ml tubes.

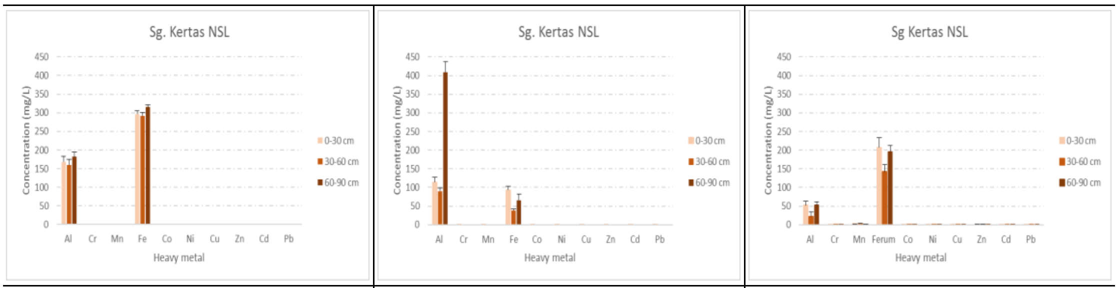
Data Analysis

All the experiments were carried out in triplicates and data presented as mean values of three independent replicates. Data were further analysed using analysis of variance (ANOVA). Statistical analysis for all experiments was performed by using SAS through factorial analysis of variance followed by Tukey's test with significant different at P<0.0001.

Results and discussion

Sungai Kertas Inert Waste non-sanitary landfill

The results in *Fig. 1* showed heavy metal concentration in an active non-sanitary landfill with a radius of 5 to 10 m, 10 to 15 m and 15 to 20 m at different depth 0 to 30 cm, 30 to 60 cm and 60 to 90 cm of Sungai Kertas non-sanitary landfill. Results of heavy metal showed that Al and Fe concentration is higher at all point of Sungai Kertas landfill.



A B C

Figure 1. *The pattern of 10 heavy metal concentration depicted Sungai Kertas non-sanitary landfill at different soil depths (0-30 cm, 30-60 cm, and 60-90 cm) and radiuses (A: 5-10 m, B: 10-15 m and C: 15-20 m) of the active landfill*

Among the analyzed heavy metals, Al had the highest concentrations up to 409.786 mg/kg within active non-sanitary soil heavy metal concentration. Compared altogether between these three radiuses of Sungai Kertas non-sanitary landfill, the heavy metal concentration at radius 5 to 10 m showed the highest result for Fe; 297.023 mg/kg, 291.954 mg/kg and 314.758 mg/kg. The lowest results showed by Al at radius 15 to 20 m; 52.089 mg/kg, 22.256 mg/kg and 53.188 mg/kg meanwhile Fe at radius 10 to 15 m showed 93.175 mg/kg, 38.714 mg/kg and 65.584 mg/kg.

Kuang Inert Waste non-sanitary landfill

The results in *Fig. 2* showed heavy metal concentration in an active non-sanitary landfill with a radius of 5 to 10 m, 10 to 15 m and 15 to 20 m at different depth 0 to 30 cm, 30 to 60 cm and 60 to 90 cm of Kuang non-sanitary landfill. Results of heavy metal showed that the highest concentration is depicted by Fe. The concentration is

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higher at the centre and the middle point of Kuang landfill. Compared altogether between these three radius of Kuang non-sanitary landfill, heavy metal concentration showed the highest result for Fe at the radius 15 to 20 m; 593.954 mg/kg, 546.171 mg/kg and 570.699 mg/kg and radius 10 to 15 m; 433.383 mg/kg, 235.407 mg/kg and 230.230 mg/kg. The lowest results showed by Al at radius 10 to 15 m; 22.311 mg/kg, 36.557 mg/kg and 32.934 mg/kg well as radius 5 to 10 m; 44.811 mg/kg, 29.436 mg/kg and 36.277 mg/kg.

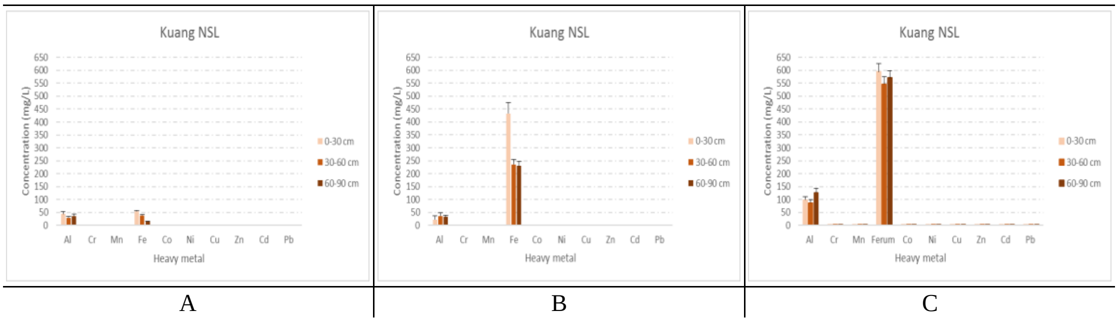

Figure 2. *The pattern of 10 heavy metal concentration depicted Kuang non-sanitary landfill at different soil depths (0-30 cm, 30-60 cm, and 60-90 cm) and radiuses (A: 5-10 m, B: 10-15 m and C: 15-20 m) of the active landfill*

Dengkil Inert Waste non-sanitary landfill

The results in *Fig. 3* showed heavy metal concentration in an active non-sanitary landfill with a radius of 5 to 10 m, 10 to 15 m and 15 to 20 m at different depth 0 to 30 cm, 30 to 60 cm and 60 to 90 cm of Dengkil non-sanitary landfill. Results of heavy metal showed that the highest concentration is depicted by Al. The concentration is higher at radius 10 to 15 m of Dengkil landfill.

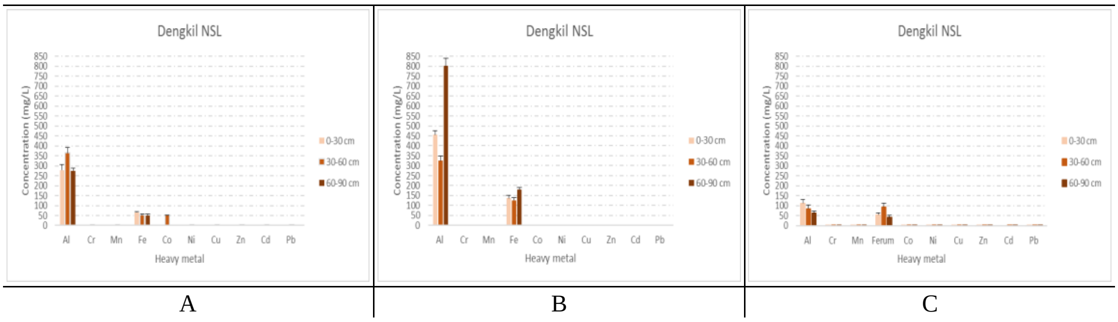

Figure 3. *The pattern of 10 heavy metal concentration depicted Dengkil non-sanitary landfill at different soil depths (0-30 cm, 30-60 cm, and 60-90 cm) and radiuses (A: 5-10 m, B: 10-15 m and C: 15-20 m) of the active landfill*

Compared altogether between these three radiuses of Dengkil non-sanitary landfill, heavy metal concentration showed the highest result for Al at radius 10 to 15 m; 455.457 mg/kg, 325.270 mg/kg and 800.804 mg/kg followed by radius 5 to 10 m; 278.700 mg/kg, 364.215 mg/kg and 275.386 mg/kg. The lowest results showed by Fe at all point. The highest concentration for Fe showed by radius 10 to 15 m;

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137.191 mg/kg, 126.688 mg/kg and 178.860 mg/kg followed by radius 15 to 20 m; 54.143 mg/kg, 93.842 mg/kg and 41.216 mg/kg then radius 5 to 10 m; 65.457 mg/kg, 52.985 mg/kg and 51.282 mg/kg. There is a concentration of Co that showed up to 52.985 mg/kg at 30-60 cm depth for radius 5 to 10 m.

Tanjung Dua Belas sanitary landfill

The results in *Fig. 4* showed heavy metal concentration in an active sanitary landfill with a radius of 5 to 10 m, 10 to 15 m and 15 to 20 m at different depth 0 to 30 cm, 30 to 60 cm and 60 to 90 cm of Tanjung Dua Belas sanitary landfill. Results of heavy metal showed that the highest concentration is depicted by Fe and Al. The concentration is higher at radius 10 to 15 m of Tanjung Dua Belas landfill. Compared altogether between these three radiuses of Tanjung Dua Belas sanitary landfill, heavy metal concentration showed the highest result for Al at radius 10 to 15 m; 85.182 mg/kg, 159.445 mg/kg and 567.264 mg/kg followed by Fe; 113.967 mg/kg, 161.552 mg/kg and 321.409 mg/kg. The radius 15 to 20 m concentration also lead by Al and Fe. Each showed 133.694 mg/kg, 120.739 mg/kg, 137.235 mg/kg for Al concentration and 170.976 mg/kg, 83.036 mg/kg and 162.968 mg/kg for Fe concentration. Meanwhile, at a radius 5 to 10 m, the concentration showed almost equal between Al and Fe. Al concentration at radius 5 to 10 m; 104.207 mg/kg, 138.692 mg/kg and 136.317 mg/kg followed by Fe; 120.165 mg/kg, 106.395 mg/kg and 103.827 mg/kg.

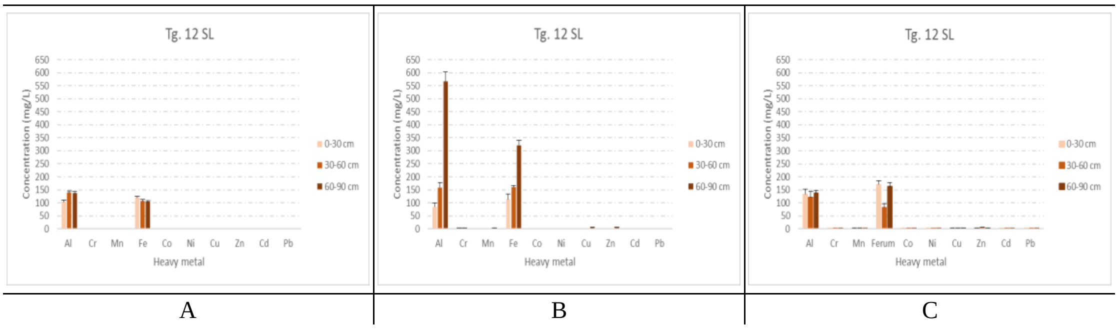

Figure 4. *The pattern of 10 heavy metal concentration depicted Tanjung Dua Belas sanitary landfill at different soil depths (0-30 cm, 30-60 cm, and 60-90 cm) and radiuses (A: 5-10 m, B: 10-15 m and C: 15-20 m) of the active landfill*

Jeram sanitary landfill

The results in *Fig. 5* showed heavy metal concentration in an active sanitary landfill with a radius of 5 to 10 m, 10 to 15 m and 15 to 20 m at different depth 0 to 30 cm, 30 to 60 cm and 60 to 90 cm of Jeram sanitary landfill. Results of heavy metal showed that the highest concentration is depicted by Fe and Al. The concentration is higher at the radius 15 to 20 m of Jeram landfill. Compared altogether between these three radiuses of Jeram sanitary landfill, heavy metal concentration showed the highest result for Al and Fe at radius 15 to 20 m; each at 219.009 mg/kg and 232.830 mg/kg for 0 to 30 cm depth. Followed by Fe at radius 10 to 15 m; 239.127 mg/kg for 30 to 60 cm depth. Meanwhile, for Al and Fe concentration at radius 5 to 10 m showed almost equal readings at all depth. Each showed 32.632 mg/kg, 82.432 mg/kg and 52.744 mg/kg for Al and 104.709 mg/kg, 168.297 mg/kg and 151.001 mg/kg for Fe.

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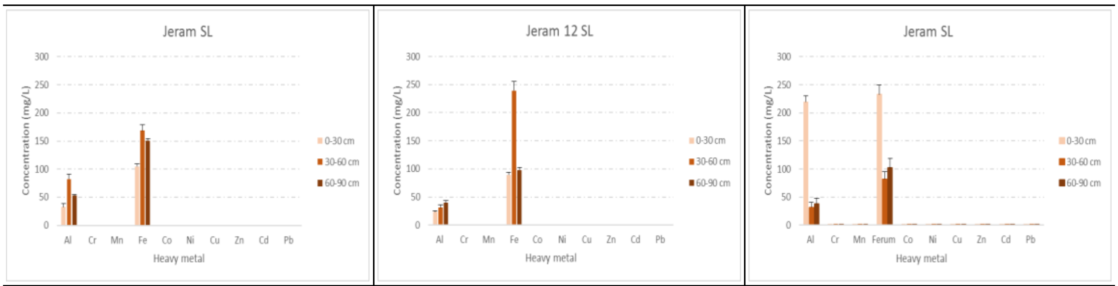
Analysis of variance confirmed the findings by exhibiting highly significant differences ($P<0.0001$) between the radius, depth, and heavy metal concentration at three different radiuses, three different depths, and ten heavy metals concentration. This clearly demonstrates that environmental factors and landfill area background can have an important influence on the accumulation of certain heavy metal and its content. Al and Fe concentration showed the highest pattern on all 4 active landfill sites. This might probably cause by the transportation of metals that may vary between relatively unpolluted systems and disturbed or polluted systems as well as the site context. Particulate transport is dominant in unpolluted systems where metal inputs are principally from the erosion of watershed substrates (Elder, 1989).



A B C

Figure 5. *The pattern of 10 heavy metal concentration depicted Jeram sanitary landfill at different soil depths (0-30 cm, 30-60 cm, and 60-90 cm) and radiuses (A: 5-10 m, B: 10-15 m and C: 15-20 m) of the active landfill*

In disturbed or polluted systems, for example, those affected by acid-mine drainage, point sources commonly deliver metals in a soluble phase or associated with organic matter, and the proportion of dissolved metals and their bioavailability tends to be higher (Solomons and Förstner, 1984). Leachate is still being produced even though most of the non-sanitary landfills in Selangor have been closed or still operating. This leachate production still occurs and badly affect the nearest water system and soil due to the location of the landfills near the water system as a source of water to the local residents. Therefore, environmental factors and landfill area background can have an important influence on the accumulation of certain heavy metal and its content. There are high contaminants levels in the groundwater underneath the non-engineered waste disposal sites in Selangor. The rate and amount of pollutant penetration through the soil is also influencing the concentration of heavy metal present in the landfill environment and finally lead to different capability depiction of heavy metal binding in the soil.

Conclusions

It can be summarised that heavy metal contents in Sungai Kertas non-sanitary landfill, Kuang non-sanitary landfill, Dengkil non-sanitary landfill, Tanjung Dua Belas sanitary landfill and Jeram sanitary landfill were accumulated by high Al and Fe. A few of the landfills had been exposed to Cr, Mn, Zn, Co, and Cu below 5.0 mg/kg. They are Sungai Kertas non-sanitary landfill, Dengkil non-sanitary landfill and Tanjung Dua Belas sanitary landfill. Kuang non-sanitary landfill and Jeram sanitary landfill showed only Al and Fe contaminations. In other words, there are particular locations as well as

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depth and radius where specific heavy metals signature accumulates with high concentration levels and some with low level. This raises the importance and needs for an effective potential model system for future research to investigate in depth the environmental factors such as the type of soil and waste that influence heavy metals profile and distribution or controlling heavy metals accumulation.

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