International Journal of Environmental Monitoring and Analysis
2013; 1(2): 71-77
Published online May 30, 2013 (http://www.sciencepublishinggroup.com/j/ijema)
doi: 10.11648/j.ijema.20130102.16

Hazards due to polycyclic aromatic hydrocarbons (PAHs) and heavy metals at the closed Kubang Badak landfill, Selangor

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To cite this article:
Nor Amani Filzah Mohd Kamil, Suhaimi Abdul-Talib. Hazards Due to Polycyclic Aromatic Hydrocarbons (PAHs) and Heavy Metals at the Closed Kubang Badak Landfill, Selangor. *International Journal of Environmental Monitoring and Analysis*. Vol. 1, No. 2, 2013, pp. 71-77. doi: 10.11648/j.ijema.20130102.16

Abstract: Demand on redevelopment of a closed landfill is increasing due to limited land resources. However, the hazards of a closed landfill remain unknown. Thus, there is a need to assess the health hazard for a closed landfill to ensure its suitability for future development. The aim of this study is to evaluate the distributions and hazards based on soil samples taken from a closed landfill located at Kubang Badak, Selangor, with specific focus on Polycyclic Aromatic Hydrocarbons (PAHs) and heavy metals. A total of ten soil samples were collected using hand auger. PAHs were extracted using Microwave Assisted Extraction (MAE) and analysed using Gas Chromatography-Mass Spectrometer, while heavy metals were digested with MAE and analysed using Inductively Coupled Plasma. Concentrations of PAHs were compared to guidelines from Canada, Netherlands and Denmark. Concentrations of heavy metals were compared to guidelines from United Kindom.. Results showed that 12 PAHs compounds were distributed consistently except for phenanthrene and anthracene. Most abundant was phenanthrene with an average value of 62.02 μg/kg. For heavy metals, Zn was found the most abundant ranging from 256.9-666.05 mg/kg. This is followed by Mn (29.05-262.5 mg/kg), V (3.85-174.65 mg/kg), Ga (34.4-182 mg/kg), Rb (63.7-135.8 mg/kg), Cr (37.8-136.85 mg/kg), Sr (0-158.2 mg/kg), Mg (16.1-173.6 mg/kg), Cu (0-201.6 mg/kg) and Pb (0.7—89.25 mg/kg). Two heavy metals, i.e. Cr and As were found to have concentrations above concentration limits permissible for residential activities. Whilst PAHs compounds were distributed uniformly, heavy metals were not. Based on the findings, landfill area is not suitable for residential area development as concentrations of Cr and As are above permissible limits.

Keywords: Closed Landfill, Heavy Metals, Health Hazard, Pahs, Soil Contamination

1. Introduction

Partially degraded municipal waste is associated with both organic and inorganic waste. Organic waste includes polycyclic aromatic hydrocarbon (PAHs), Polychlorinated Biphenyls (PCB), Polychlorinated Dibenzo-p-dioxins (PCDD) and Polychlorinated Dibenzofurans (PCDF). Inorganic wastes includes heavy metals, nitrogen, nitrate, ammonia, phosphorous and phosphate. These pollutants originate from waste materials such as oils, pesticides, discarded electronic equipment, debris waste, used paints, organic waste, etc. These wastes must be properly managed to minimize their impacts on the environment and human health. In Malaysia, these wastes are mostly dumped into landfill area [1]. Dumping activities at landfills make it a major reservoir and sink for pollutants because of the high soil holding capacity. In addition, soil also has the capability to accumulate the pollutants. Soil is considered as a good indicator of pollution and environmental hazard [2].

Land used for waste disposal may be contaminated. Its redevelopment often requires that unacceptable risks are assessed and managed so that the site becomes suitable for its new use. One of the major criteria to be fulfilled before such land can be re used is to ensure that pollutant levels are within the clean-up guidelines. This is to ensure that the site is safe with no health hazard to human and the environment.

Micropollutants such as PAHs and heavy metals are considered among the most adverse pollutants. PAHs are ubiquitous contaminants and highly toxic, mutagenic and carcinogenic that can be linked to other health problem which had attracted the interest of the research community [3,4]. Sixteen PAHs compound have been identified by USEPA as priority pollutant. They represent the most adverse effects that are caused be anthropogenic activities [5]. Ingestion, inhalation and skin contact of heavy metals on humans can cause disturbance to biological reactions, long lasting harm to vital organs or even death [6].

Therefore, the investigation of soil pollutants at closed landfill is needed to ensure the landfill is safe for its future development and subsequent uses. Thus, the objective of this study was to evaluate the distributions and health hazards of soil samples from a closed landfill located at Kubang Badak, Selangor, with focusing on PAHs and heavy metals.

2. Experimental

2.1. Sampling Site

Kubang Badak landfill in Batang Berjuntai, Selangor is located in the midwestern part of the Malaysian Peninsular. The site was properly closed by the authority on 7th April 2007 and was replaced with Bukit Tagar sanitary landfill, which is located near the site. The closed landfill site covered an area of 1 hectare. The servive area for the Kubang Badak landfill extend from Tanjong Karang to Sepang covering more than 70% of Selangor state [7]. A large number of steel related industries, cement industries, wood industries are located in the servive area. In addition, there are also residential areas and a university (UNISEL) located in the area [8]. The closed landfill is located next to Sungai Selangor. Leachate produced from the site can easily pollute the river. The sampling sites were chosen so as to represent the pollution distribution on the site.

Soil samples were collected from ten locations during the period of November 2009. For each location, duplicate samples were collected from approximately 1m2 areas. Sampling points are shown in Figure 1 and details of these sampling locations are provided in Table 1.

Table 1. *Description of sampling station at Kubang Badak landfill*

Sampling Station	Description
S1	Upper level, covered with thick layer of soil, near to mobile flare
S2 to S6	Upper level, covered with thick layer of soil
S7	Lower level, covered with thin layer of soil
S8 to S9	Middle of upper and lower level, covered with soil
S10	Lower level, near to palm trees area

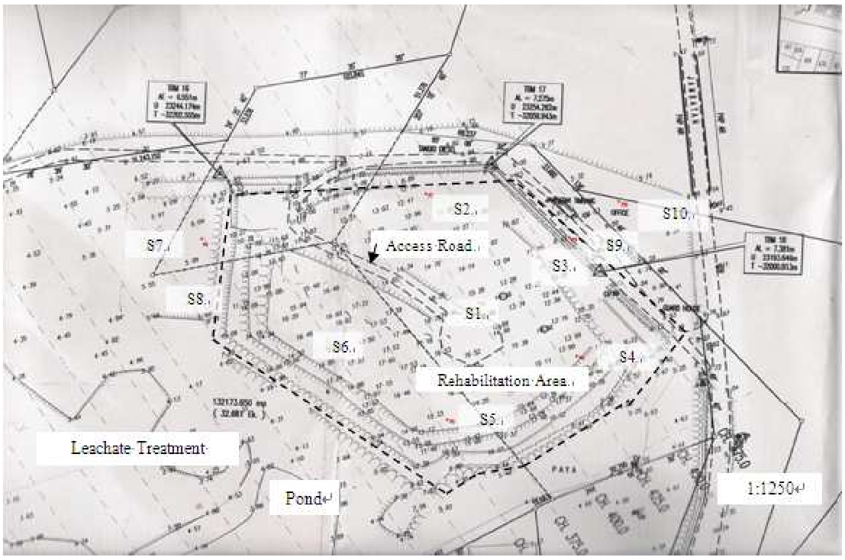

Figure 1. *Location map of the sampling stations in Kubang Badak landfill.*

2.2. Sampling Site and Preparation

Landfill soil samples were collected using a hand auger at depths of 1m and kept in high-density polyethylene (HDPE) plastic bag. All samples were transported to laboratory in an icebox to minimize sample degradation and to avoid loss of PAHs through volatilisation. In preparation for analysis, samples were air dried at room temperature and sieved to collect particles less a 1mm. The selected size of particles was based on [9], which reported that PAHs are more abundant in finer particles. The particles were stored in plastic bags and sealed, homogenized and kept in this manner until ready for analysis.

2.3. PAHs Extraction and Analysis

For sample analysis, 500 mg of the contaminated sand sample was dissolved in 25 ml of n-hexane and acetone 7:3

(v/v). The extractions were performed with the pressurized microwave extraction system (MAE) Multiwave 3000 (Rotor 8XF100 SOLV and solvent safety system under controlled pressure for a specified time. Extraction process was done within 40 minutes under a pressure of 10 bars. When the extraction period was completed, the equipment was allowed to cool down to room temperature for 20 minutes. Subsequently, the samples were filtered with Whatman glass fibre filters with pore size of 11 µm and kept in 25 ml universal bottles. The samples were concentrated by means of a rotary evaporator to about 1 ml.

PAHs concentrations were measured using Gas Chromatography Mass Spectrometer (Perkin Elmer Clarus 600), equipped with Elite Column 5MS with 30m long X 0.25mm internal dimension X 0.25µm thickness. The injector was operated at 250oC in the splitless mode with a 3 minute splitless period. Helium was used as the carrier gas with 1 ml/min constant flow rate. The column temperature was initially set at 50oC for 1 min, increased to 250oC at a rate of 25oC/min and then kept constant at 30 minutes. The total run for each sample is set to 54 minutes.

2.4. Heavy Metals Digestion and Analysis

100 mg of landfill soil was digested with 2ml of HNO3 (65%), 1ml HCl (37%), 0.5ml HF (40%) and 0.5ml H3PO4 (85%). Digestion was performed with the pressurized microwave extraction s ystem (MAE) Multiwave 3000 (Rotor 8XF100 SOLV and solvent safety system) controlled power for a specified time as shows in Table 2. Temperature and pressure reaction conditions were set to 218°C and 30 bars, respectively. The resulting solution was transferred into measuring flask to a volume of 70 mL. Subsequently, the samples were filtered with Whatman glass fibre filters with pore size of 0.45 µm. For 5ml final digestion, 100 µL of internal standards, i.e., 2ppm Sc was added.

Table 2. *Multiwave Program for Heavy Metals Digestion*

Power (W)	Time (min)	Fan
400	6	0
1000	20	0
0	15	3

Table 4. *Concentration limit (mg/kg) for Heavy Metals.*

HM	As	Pb	Cu	Cr	Zn
CLEA[a]	20	450	NA	130	NA
CLEA[b]	NA	750	NA	5000	NA

[a]Residential[b]Commercial and Industrial

Heavy metals concentration was measured by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES, Perkin Elmer Optima 7000). The calibration of the instrument for each metal was performed according to its wavelength and standard solutions. Multi heavy metals standard solutions were prepared at 0.01ppm, 0.1ppm, 1ppm and 10ppm.

2.5. Comparison with Health Hazard Guidelines

Health hazard guideline values for PAHs in soil from three countries are shows in Table 3. These guidelines provides useful indicator values for the purpose of estimating the potential hazard to human health [10,11,12]. Health hazard guideline for heavy metals in soil as stipulated in United Kingdom [13] is shown in Table 4, which covers three categories, i.e., residential, commercial and industry [13].

Table 3. *Soil Quality Criteria (µg/kg) for PAHs from Netherlands, Denmark and Canada*

Country	Na	Ph	An	Fl	Py	BaA	Ch	Bkf	BaP	IP	DaH	BgP	PAHs (sum)
Canada	600	5000	NA		10000	1000	NA	1000	700	1000	1000	NA	NA
Netherlands	NA								100	NA	100	NA	1500[a]
Denmark	NA												1000[b]

[a]Sum of 5 PAHs: Fluotherene, Benzo(b+k)fluoranthene, Benzo(a)pyrene, Dibenz(a,h)anthracene, Indeno(1,2,3-cd)pyrene.
[b]Sum of 10 PAHs: Napthalene, Phenanthrene, Anthracene, Fluoranthene, Benz(a)anthracene, Chrysene, Benzo(b+k)fluoranthene, Benzo(a)pyrene, Indeno(1,2,3-cd)pyrene, Benzo[ghi]perylene.

Table 5. *Concentration of PAHs in Kubang Badak landfill (µg/kg)*

PAHs	Na	Flu	Ac	Ph	An	Fl	Py	BaA	Tr	BjF	BaP	BgP
AVE	16.41	15.04	15.33	62.02	23.01	15.70	15.53	16.53	15.59	16.30	16.50	19.98
STDEV	0.054	0.160	0.004	43.159	9.182	0.040	0.266	0.006	0.054	0.009	0.009	0.021
MIN	16.34	14.95	15.32	15.64	16.29	15.66	15.24	16.52	15.56	16.29	16.4	19.96
MAX	16.51	15.48	15.33	121.5	43.92	15.79	15.83	16.54	15.73	16.32	16.52	20.02

Na: Napthalene, Ph: Phenanthrene, An: Anthracene, Fl: Fluoranthene, Py: Pyrene, BaA: Benz(a)anthracene, Ch: Chrysene, BkF: Benzo(b+k)fluoranthene, BaP: Benzo(a)pyrene, IP: Indeno(1,2,3-cd)pyrene, DaH: Dibenz(a,h)anthracene, BgP: Benzo[ghi]perylene

3. Result and Discussion

3.1. Distribution of PAHs Concentration

Based on this study, twelve PAHs were found to be present in the samples. These PAHs were 2-3 rings, i.e. naphthalene, fluorene, acenaphthene, phenanthrene and anthracene, 4-5 rings, i.e. fluoranthene, pyrene, benz(a)anthracene, triphenylene, benzo(j)fluoranthene, benzo(a)pyrene and 6 rings, i.e. Benzo(g,h,i)perylene. Tables 5 shows that all the PAHs compounds detected were distributed consistently except for phenanthrene (stdev: 43.16) and anthracene (stdev: 9.18).

For the individual compound distribution, phenanthrene is the most abundant PAHs in soil samples. Phenanthrene was also reported to be the most abundant in leachate, river and groundwater close to landfill sites in Ulu Maasop landfill, Kuala Pilah, Negeri Sembilan and Taman Beringin landfill, Selangor [14]. This could be attributed to the fresh input of phenanthrene to landfill through illegal dumping of coal tar. Some of lower molecular weight PAHs, e.g. naphthalene, fluorene and acenaphthene are more volatile and have higher bioavailability compared to phenanthrene and anthracene. Therefore, the lower molecular weight PAHs show low concentrations in the analysed samples. Furthermore, higher molecular weight PAHs, i.e. Benz(a)anthracene, Triphenylene, benzo(j)fluoranthene, Benzo(a)pyrene and BgP: Benzo[ghi]perylene, were found predominant in smaller particles which deposit slowly from the atmosphere.

The presence of phenanthrene and anthracene in high concentrations is an indicator of fresh input of PAHs in soil.

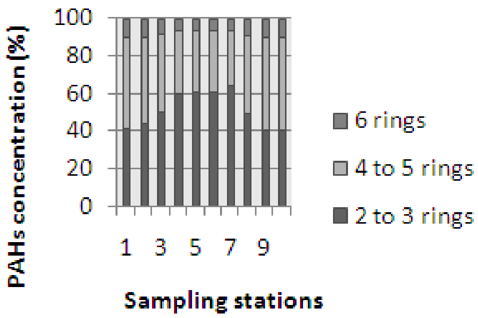

Figure 2. *Distribution of PAHs in different sampling locations.*

At most sampling stations, 2-3 rings PAHs were predominant, accounting for around 40 – 65% to the total PAHs, followed by 4-5 rings PAHs accounting for 30 – 50% and only 6 – 10% for 6 rings PAHs as tabulated in Figure 2. The distribution of PAHs was closely similar to the distribution of PAHs found in landfill Tagarades, Greece [15]. Higher concentrations of 2-3 rings PAHs in soil may be due 2-3 rings PAHs being more hydrophilic than 4-6 rings PAHs, thus 2-3 rings tend to dissolve in water and wet deposition process from atmosphere are higher compare to 4-6 rings PAHs [9].

The maximum value of total PAHs concentration was found to be 329.13µg/kg much lower than those measured in Tagarades waste landfill, Thessalonik, Greece [15], restored landfill, Hong Kong [2], Kouroupitos waste disposal site, Crete, Greece [16] and Marka landfill, Jordan [17] as shown in Table 6. Highest concentration found at Tagarades waste landfill [15] and Marka landfill [17] was influenced by open combustion and smouldering process. From the investigation of the landfill site, no large open combustion process was found. The landfill had only experienced several small fires in the past, probably due to spontaneous ignition of methane released from mobile flare at S1.

3.3. Hazard Assessment on PAHs

Individual PAHs were compared to two guidelines from Canada and Netherlands [12,10] as shown in Table 7. All PAHs compounds detected were below the permissible concentration limits. Although PAHs level was detected below the guideline limits for residential uses, it is not recommended for housing development especially houses on land surface, e.g. terrace and bungalow. Phenanthrene may contribute health hazard to residents through ingestion of edible plants. Fismes et al.,[18] had reported significant concentrations of PAHs in plant, e.g. potato, lettuce and carrot. In addition, concentration of PAHs in plants can increase with increasing PAHs concentrations in soil. Reference [18] also found higher concentration in lettuce roots compared to leaves. Thus, human ingesting plant from phenanthrene contaminated soil will be exposed to hazards from phenanthrene, e.g. kidney neoplasm and skin irritation.

Table 7. *Comparison of Individual PAHs with Guideline (µg/kg)*

PAHs	Na	Flu	Ac	Ph	An	Fl	Py	BaA	Tr	BjF	BaP	BgP
MAX	16.51	15.48	15.33	121.5	43.92	15.79	15.83	16.54	15.73	16.32	16.52	20.02
Guid[a]	600	NA	NA	5000	NA	NA	10000	1000	NA	NA	700	NA
Guid[b]	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	100	NA

Na: Naphthalene, Flu: Fluorene, Ac: Acenaphthene, Ph: Phenanthrene, An: Anthracene, Fl: Fluoranthene, Py: Pyrene, BaA: Benz(a)anthracene, Tr: Triphenylene, BjF: benzo(j)fluoranthene, BaP: Benzo(a)pyrene, BgP: Benzo[ghi]perylene
Guid[a]: Guideline from Environment Canada, 2003 for residential uses
Guid[b]: Guideline from Ministry of Housing Spatial Planning and Environment, 2000

Table 9. *Concentration of heavy metals in Kubang Badak landfill(mg/kg)*

HM	As	Pb	Cu	Mg	Sr	Cr	Rb	Ga	V	Mn	Zn
AVE	7.81	26.29	38.12	58.80	71.96	89.81	98.84	115.26	105.84	121.42	406.21
STDEV	8.97	28.56	60.17	48.58	53.88	36.15	21.57	50.86	55.18	71.74	118.52
MIN	0	0.7	0	16.1	0	37.8	63.7	34.3	3.85	29.05	256.9
MAX	26.9	89.25	201.6	173.6	158.2	136.85	135.8	182	174.65	262.5	666.05

However, the Kubang Badak landfill site is suitable for commercial and industrial uses. Human can be exposed to pollutants through air inhalation, dermal absorption and ingestion [19]. Concentrations of phenanthrene detected in this study are acceptable and will not cause health hazards to human through air inhalation and dermal absorption.

Total PAHs concentrations were compared to recommended levels imposed by soil clean-up guidelines from Netherlands and Denmark [10, 11] as shown in Table 8. The total PAHs found in this study were below the recommended values and thus, the site is acceptable for human activities. However, the site is not recommended for residential area due to potential of contamination along the food chain as a result of agricultural activities.

Table 8. *Comparison of Total PAHs with Soil Clean-up Guidelines*

Country	Sum of 5 PAHs[a]	Sum of 10 PAHs[b]
This study	32.32	250.8
Netherlands	1500[a]	
Denmark		1000[b]

Table 6. *Literature data concerning concentrations of PAHs in landfill soils*

Researchers	Total PAHs (µg/kg)	Location
This study	194.59 – 329.13	closed landfill, Selangor, Malaysia
Ref. [15]	11.2 – 1475	operating landfill, Thessaloniki, Greece
Ref. [2]	380.1	restored landfill, Hong Kong
Ref. [16]	16 – 775	operating landfill, Crete, Greece
Ref. [17]	33 – 3560	closed landfill, Marka Amman, operating landfill, Ekeeder/Irbid, Jordan

3.2. Distribution of Heavy Metals Concentration

The concentration of heavy metals in soil samples collected within Kubang Badak landfill are shown in Table 9. The highest mean concentration was noted for Zinc (Zn),followed by Manganese (Mn), Gallium (Ga), Vanadium (V), Rubidium (Rb), Chromium (Cr), Barium (Ba), Strontium (Sr), Magnesium (Mg), Copper (Cu), Plumbum (Pb) and Arsenic (As).

Normal concentration range for Zn in soil is between 1-400mg/kg [20]. Zn concentration in soil samples detected in this study is below than 400mg/kg except for S1 (406mg/kg), S5 (666.05mg/kg), S7 (418.25mg/kg) and S10 (531.3mg/kg). The most abundant metal after Zn was Mn, which varies from 29.05 to 262.5mg/kg. This is most likely due to the contamination from steel industry nearby which produces scrap iron and used to dispose their waste to the landfill. Same as Mn, V also one of component use steel production speciality in steel alloy. Cr concentration varies from 37.8 to 136.85 mg/kg. A cement industry located nearby to the landfill site used to dispose their waste from the linings for the rotaries containing Cr. These waste might caused Cr emissions due to their wear and friction. Normal Cu concentration range in soil is below 20mg/kg [20]. Cu concentration range in this study was 0 – 201.6mg/kg and this shows that the investigated area was highly polluted with Cu. Cu is commonly used as timber preservative in wood industries. During site investigation, wood factory was found close to landfill site and this could be the reason for high concentration of Cu. Pb concentration range detected in this study was 0.7-89.25mg/kg.

Table 10. *Literature data concerning concentrations of heavy metals in landfill soils*

Researchers	Location	Pb		Cr		Cu		Zn	
		min	max	min	max	min	max	min	max
This study	Closed landfill, Kubang Badak, Selangor, Malaysia	0.7	89.25	37.8	136.85	0	201.6	256.9	666.05
Ref. [25]	Closed landfill, Likeng village, Guangzhou	10.1	82.7	15.8	702.6	NM		17.4	247.1
Ref. [21]	Sanitary landfill, Sg. Kembong, Selangor, Malaysia	3.2	78	4	76	2	326.5	12.4	128.4
Ref. [24]	Closed unlined landfill, Efkarpia, Thessaloniki	2.50	92.5	3.88	171.88	8.13	356.25	6.38	343.75
Ref. [17]	Close landfill, Marka Amman, operating landfill, Ekeeder/Irbid, Jordan	35	2380	35	142	NM		NM	
Ref. [20]	*Normal concentration in soil	NA	NA	NA	NA	0	20	1	400

NM: Not Measured
Unit: mg/kg

Table 11. *Comparison of heavy metals (mg/kg) with CLEA, 2002*

HM	As	Pb	Cu	Mg	Sr	Cr	Rb	Ga	V	Mn	Zn
MAX	26.9	89.25	201.6	173.6	158.2	136.85	135.8	182	174.65	262.5	666.05
CLEA[a]	20	450	NA	NA	NA	130	NA	NA	NA	NA	NA
CLEA[b]	NA	750	NA	NA	NA	5000	NA	NA	NA	NA	NA

Range of concentrations for Cr, Pb, Zn and Cu that determined in this study are quite similar reported from other studies as shown in Table 10. Maximum concentrations for Zn, Cu and Cr were detected more than 100mg/kg except for study by [21]. Whereas, maximum concentrations for Pb was detected below than 100mg/kg except for study conducted by [17]. These concentrations ranges are very important for bioremediation purpose, as high concentrations of metals can retard degradation of organic contaminant. References [22] reported that high concentrations of Cu caused an incomplete mineralization of phenanthrene by soil microorganisms. In contrast, low concentrations are essential for the function of certain enzymes in microorganisms for growth, e.g. Cu and Zn, these are referred as trace elements [23].

4. Hazard Assessment on Heavy Metals

Zn and Mn concentrations were detected in high concentration in this study. When humans are exposed to high concentration of Zn and Mn, it will decrease the oxygen level in cells, which Cu acts as catalyst in formation of hemoglobin [26]. V is considered toxic by National Institute for Occupational and Health (NIOSH) which cautioned that 35 mg/m3 can cause permanent health problems or death [27]. Only two sampling stations, i. e. S7 (136.85mg /kg) and S8 (130.2mg/ kg) for Cr were above 130mg/ kg, which is the concentration limit for residential area. High concentration of Cr can lead to DNA damage [28]. Acute toxicity for Cr ranges between 50 to 150µg/kg High Cr concentration in human body, can damage liver, kidney and blood cells through oxidations reactions [29]. Pb concentration range detected in this study was 0.7-89.25mg/kg. The concentrations are below the stipulated concentrations limit from United Kingdom, thus it shows that the investigate area Kubang Badak landfill is less polluted with Pb. Most sampling station show As is below than 20mg/kg except for S7 (26.95mg/kg). As a result, the site is not suitable for residential area.

Several heavy metals, i.e. Cr and As were found to have concentration above concentration limits for residential as shown in Table 11. These metals can be transferred to human body through ingestion from edible plants. Reference [30] has reported positive relationship between leaf and Zn concentration in soil. All these metals must below than concentration limits by means of remediation process before planning for residential area. Although, the site is not suitable for residential, the site can be used for commercial and industry.

5. Conclusion

All the PAHs compounds were distributed uniformly except for phenanthrene and anthracene. Heavy metals on the other hand were not distributed uniformly where Zn shows high concentrations. Referring to the guideline from Netherlands, Denmark and Canada [10,11,12] for PAHs, from United States [13] for heavy metals, the landfill area was not suitable for residential area. Cr and As are above than concentrations limits for residential uses. These pollutants constitute health hazard to human through ingestion of edible plant. However, the landfill area is suitable for commercial and industry sectors. It is recommended that this preliminary investigation be further developed, to include sampling on nearby river, Sungai Selangor to identify the hazard effect of landfill site to the aquatic environment.

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