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Transformation and Transport Phenomena of Carcinogenic Micro-Pollutants from Municipal Landfill

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Abstract: Municipal landfills produce toxic leachate that contains micro-pollutants especially polycyclic aromatic hydrocarbons (PAH) and endocrine descriptive chemicals (EDC). These pollutants are bioactive, carcinogenic and persistent in environment that tend to accumulate and cause detrimental effects to the ecosystem, food chains and human health**.** In this study, Papan landfill that is located in between the towns of Papan and Lahat, Perak was selected to monitor the discharge of leachate starting from July 2018. Water samples were collected from 9 sampling points included from landfill and along Johan river that flows to Kinta river which is the principal source of drinking and irrigation water in Perak. Results showed that WQI ranged from 44.18 to 86.13 at all sampling points and the highest PAHs concentration recorded was fluoranthene with 666.65ppb in treated leachate in August.

Keywords: *municipal solid waste, leachate, water quality index, polycyclic aromatic hydrocarbons, endocrine descriptive chemicals.*

1. Introduction

Diverse waste consists of municipal solid waste (MSW) which originated from different sources such as commercial, municipal services, residential, or agriculture is highly dependent on the size of population, economic development, industrialization rate and style of practices in the region [1]. When the population size increase, human activities increases that contributed to larger waste generation. Malaysian population has been increasing at a rate of 2.4% per annum or about 600,000 per annum since 1994 which contribute to the larger MSW generation [2]. In Peninsular Malaysia, about 17,000 tonnes of MSW are generated per day (6.2 million tonnes/year) in 2002 and the per capita varies between 0.5 and 0.8 kg/day and has increased to 1.7 kg/day in big cities [2-4]. This rate can lead to MSW generation of 31,000 tonnes per day in 2020 [5]. MSW comprises around twenty different categories namely wood waste, food waste, fruit waste, green waste, paper, metals, baby diapers, new print, batteries, glass, construction waste and others. These categories of MSW can be classified into organic and inorganic waste, around 80% of the total MSW will end up in landfills [6]. At present, land filling is the most usable practice for waste disposal compared to incineration and other waste disposal techniques as it has lower cost of maintenance and operation. However, leachate produced at landfills which undergo several phases such as aerobic, anaerobic methanogenic phase and finally the stabilising methanogenic phase [7] contain more than 200 organic compounds [8] that potential to cause harm to the environment and human health [9]. Among them, micro-pollutants such as PAH and EDC were found to be carcinogenic and posed harm to environment, or even human health. Sixteen (16) PAHs are in fact considered as priority pollutants by the European Union environmental protection agency, US EPA and WHO; these being BaA (benz[a]anthracene) (4-ring), Chr (chrysene) (4-ring), BbF (benzo[b]fluoranthene) (5-ring), BkF (benzo[k]fluoranthene)(5ring),BaP(benzo[a]pyrene)(5ring),DahA(dibenz[a,h]anthracene) (5-ring), IcdP (indeno[1,2,3-cd]pyrene) (6-ring), Nap (naphthalene) (2-ring), Acy (acenaphthylene) (3-ring), Ace (acenaphthene) (3-ring), Flo (fluorene) (3-ring),Phe(phenanthrene)(3-ring),Ant (anthracene) (3-ring), Fla (fluoranthene) (4-ring), Pyr (pyrene) (4-ring), and BghiP (benzo[g,h,i]perylene) (6-ring) whereas EDC include dimethyl-phthalate, diethyl-phthalate, dibuthyl-phthalate, benzyl butyl phathalate, bis(2-ethylhexyl)3phths and di-n-octyl-phathalate. These micro-pollutants tend to be bioaccumulated in organisms and biomagnified through the food chain which pose acute and chronic

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toxicity [10] such as lung cancer, bronchitis, asthma, and heart diseases [11]. Thus, this study aimed to assess the effect of municipal solid waste leachate from landfill on river water quality by using water quality index (WQI) and the occurrence and distributions of PAH and EDC from landfill by experiment and MATLAB software modelling.

2. Materials and Methods

2.1 Study area

Papan landfill as in Figure 1 is located 1.7 km within the buffer zone of the Asian Rare Earth (ARE) radioactive depository. It has an area of 10 acres land, 226.73 hectares larger compared to the old landfill Bercham which was only 96 acres in size. Papan landfill serves more capacity to receive dumps at a rate of 800 tonnes a day and the new site has 53,516 residents and 34 housing estates within a 5 km radius that projected to last for 35 years [12].

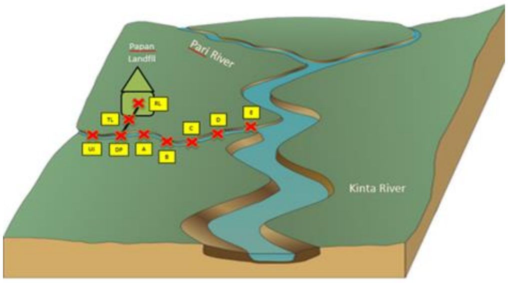

Figure 1 Papan landfill with sampling points.

2.2. Sample collection

Leachate samples from Papan landfill are collected and analysed for PAHs and EDCs. There are 9 sampling points that will be collected twice in a month for a year where 5 of them are in Papan landfill and 4 of the rest are collected along Johan river. pH, dissolved oxygen and temperature are recorded on the spot once water samples are collected. Samples are kept in a cold room at 4◦C. Parameters such as total suspended solid (TSS), chemical oxygen demand (COD), biochemical oxygen demand (BOD) and ammoniacal nitrogen (AN) are determined to obtain water quality index (WQI) of the river.

2.3. Physico-chemical parameters

2.3.1 Total suspended solids (TSS)

Aluminium pans are labelled, filter papers are cleaned and placed into the aluminium pans and put into the oven overnight for 105◦C. Heated aluminium pans with filter papers are taken out from oven after 1 day and placed into desiccator to cool down. The weight of cooled down filter paper and aluminium pan are weighted and recorded. Distilled water is used to clean the filter pump and filter paper is placed on top of the filter holder. 200ml of water samples are measured using measuring cylinder and pour them into the filter pump. The experiment is repeated with other 8 collected water samples. Filter pump is on while distilled water can be added to the surrounding of the pump to ensure that the pump is clear from suspended solids. Once all the water samples have been filtered, watch glass is removed and filter paper containing the suspended solids are placed in the aluminium pan. The pan with filter papers are put into the drying oven at 105˚C for 1 hour. After one hour, the heated aluminium pan containing the filter papers are taken out from the drying oven and placed in a desiccator for 10 minutes to cool down to room temperature. Aluminium pan together with the filter paper containing suspended solids are weighted and recorded. TSS in mg/L are calculated as the below equation.

$$TSS = \frac{\text{Final reading- Initial reading}}{\text{Volume of sample}} \quad (1)$$

2.3.2 Ammoniacal nitrogen

25 mL of water samples are measured using measuring cylinder and placed into a conical flask. For raw leachate, dilution is required in the ratio of 1:500 using volumetric flask and placed the 25mL of diluted raw leachate into conical flask. 3 drops of mineral stabilizer, 3 drops of polyvinyl alcohol and 1 ml of Nessler reagent are added to the water samples in the conical flask. The water samples allowed to mix well for 1 minute.10 mL of the mixed water sample is measured from conical flask and placed into the cuvette. Zero calibration is done by using distilled water that set as blank. Water samples filled in cuvette are inserted into lab spectrophotometer to observe and reading are recorded.

2.3.3 Chemical oxygen demand

The COD reactor is pre-heated until the temperature raised to 150◦C. 2 mL of water samples are filtered, measured and poured into the COD vial containing potassium dichromate. For raw leachate, dilution is required in the ratio of 1:500 using volumetric flask and placed 2 mL of diluted raw leachate into the COD vial. The COD vial containing water samples is then shake properly until heat is produced which indicate the exothermic process. 2ml of distilled water added to COD vial which set as the blank. All the COD vial containing water samples and blank were insert into the COD reactor and left for reaction occur for 2 hours. After 2 hours, COD vials were let to cool down for 1 hour and readings are recorded by using spectrophotometer.

2.3.4 Biochemical oxygen demand

150 mL of water samples are measured using a measuring cylinder and pour into the BOD bottle. For raw leachate, dilution is required in the ratio of 1:500 using volumetric flask and placed the 150 mL of diluted raw leachate into the BOD bottle. Blank is prepared by filling the BOD bottle with 150ml distilled water. A small amount of nitrification inhibitor was added to the BOD bottles containing water samples. 150mL of aerated distilled water was added to all BOD bottles and the initial DO for each water sample was measured by the DO probe that was equipped with a stirring mechanism. BOD bottles are then been caped well, tight and ensure that there are no bubbles inside. The BOD bottles were covered with aluminium foil and placed in the refrigerator at 20 °C temperature and left for 5 days. Final DO reading is measured by DO probe after 5-day incubation period. DO readings were recorded. BOD readings in mg/L are calculated as the equation below

$$\frac{\text{Initial reading- Final reading}}{\text{Volume of sample}} \times \text{dilution factor} \tag{2}$$

2.3.5 Extraction of PAHs and EDCs

100 mL of water sample were measured and added into a 100mL separating funnel. Dichloromethane (DCM) with a volume of 25mL was added into the same separating funnel containing with the sample. Then the mixture was shaken vigorously to release the gas until no more gas was present in the mixture. The separating funnel was left at rest for a few minutes until two layers of liquid were observed. The lower layer was extracted carefully to a conical flask. The conical flask was cover with aluminium foil to ensure no diffusion of mixture. The experiment was repeated twice with 12.5 mL of DCM. The extracted sample were then added with sodium sulphate to absorb the water until a clearer solution observed. The samples were then filtered with filter paper into a conical flask. The filtered samples are transferred to a rotary evaporator for evaporate until the volume of samples reduced to 2 mL. The samples are placed to a GCMS vial. 1 mL acetonitrile were added into the vial containing filtered sample. Vials were sealed well and covered with aluminium foil and sent to GC-MS to determine the concentration of PAH and EDC contains in the samples.

2.4. *Water quality index (WQI)*

WQI can be represented in an equation in the form of index number to indicate the quality of river water [13]. The calculation and formula of WQI is as shown in Eq (3). The range of water quality index should be $0 \leqslant \text{WQI} \leqslant 100$ where the corresponding status of water quality and their possible uses are summarized in Table 1.

$$\text{WQI} = 0.22\ \text{SIDO} + 0.19\ \text{SIBOD} + 0.16\ \text{SICOD} + 0.15\ \text{SIAN} + 0.16\ \text{SISS} + 0.12\ \text{SIpH} \tag{3}$$

Where:

SIDO = SubIndex DO (% saturation)
SIBOD = SubIndex BOD
SICOD = SubIndex COD
SIAN = SubIndex NH_3–N
SISS = SubIndex SS
SIpH = SubIndex pH

Table 1 WQI and corresponding water quality status [17]

Class	**WQI**	**Possible usage**
I	93 and above	Safe for direct drinking
II	From 78 to < 93	Requires treatment for drinking purposes
III	From 52 to < 78	Intensive treatment for drinking
IV	From 31 to <52	Only suitable for plant and domestic animal uses
V	From 11 to < 31	Cannot be used for the purposes listed in Classes I–IV

3. RESULTS AND DISCUSSIONS

3.1 Water Quality Index

Tables 2 and 3 are the WQI for both batch of samplings which illustrated in Figure 2.

Table 2 Monthly WQI first batch

Sampling point	WQI		
	July	Aug	Sept
Upstream	85.40	82.92	77.15
Discharge point	56.09	75.51	80.49
Downstream (A)	86.13	83.93	77.55
Downstream (B)	85.62	76.01	67.46
Downstream (C)	63.51	46.32	65.62
Downstream (D)	75.00	56.42	68.78
Downstream (E)	73.77	57.56	56.51

Table 3 Monthly WQI second batch

Sampling point	WQI		
	July	Aug	Sept
Downstream (B)	72.58	60.84	71.36
Downstream (C)	56.24	50.26	68.86
Downstream (D)	66.34	44.18	65.06
Downstream (E)	61.55	59.85	59.40

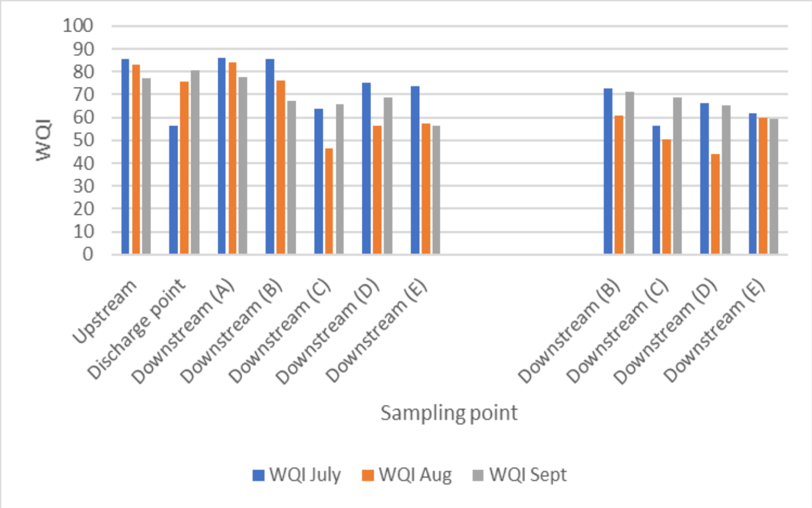

Figure 2 Monthly WQI of Johan river

The results obtained from physico-chemical analysis of water samples from different sampling locations are used to determine water quality index (WQI) as shown in Figure 2. From the results obtained, overall mostly WQI are in the range of 44.18 to 86.13 which is in the class of II and III where treatment and intensive treatment required for drinking purposes. Trend of monthly WQI fluctuated for both batches of sampling along the river which can be due to that precipitation and heavy rainfall especially during rainy season from August to November with thunderstorm [14] can increase the level of river water and enhance dilution thus effects the pollutions. However, it is noted that second batch sampling has lower WQI compared to first sampling where the lowest was in downstream D with WQI 44.18 in August recorded. This means that it is only suitable for plant and domestic animal uses. Besides, agricultural runoff and industrial discharge during batch 2 can also lead to the deterioration of the water quality.

3.2. Raw and treated leachate

The physico- chemical parameters for raw and treated leachate are showed in Figure 3. It is noted that treated leachate has lower BOD, COD, AN and TSS compared to raw leachate.

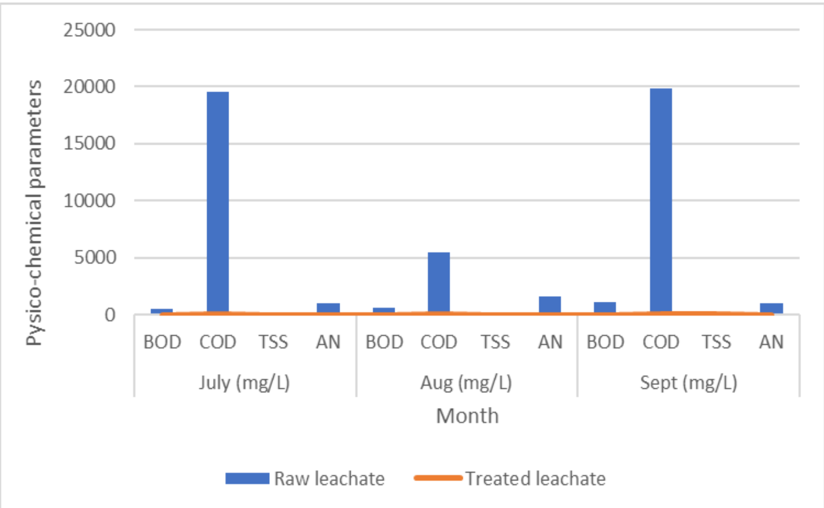

Figure 3 Raw and treated leachate

3. Polycyclic aromatic hydrocarbon (PAHs)

The monthly occurrence and distributions of PAHs along the sampling points are showed in Figures 4 to 7. Figures 4 and 5 showed the monthly PAHs taken from Papan landfill whereas Figures 6 and 7 showed the monthly PAHs taken along Johan river. It was observed that PAHs concentration in August (Figure

4), are higher in treated leachate such as fluoranthene with 666.85ppb and pyrene with 628.62ppb. Meanwhile in upstream, several micro-pollutants such as Benzo (a) anthracene, Chrysene, Benzo (b) fluoranthene, Benzo (k) fluoranthene, Benzo (a) pyrene, Indeno (1,2,3-cd) pyrene, Dibenzo (a,h) anthracene and Benzo (g,h,i) perylene are relatively higher compared to the others points. However, in September (Figure 5), point downstream D appeared to be higher in Fluoranthene, Pyrene, Benzo (a) anthracene, Chrysene, Benzo (b) fluoranthene, Benzo (k) fluoranthene, Benzo (a) pyrene, Indeno (1,2,3-cd) pyrene and Dibenzo (a,h) anthracene. Fluoranthene concentration was recorded the highest with a value of 267.23 ppb. It was supported that PAHs with lower molecular weight can happen to have lower concentrations, fluoranthene can be present in higher concentration when compared to other PAHs [15] Moreover, manufacture of agrochemicals, dyes and pharmaceuticals [16] can contribute to the higher concentration of fluoranthene where in this study some of the sampling points are taken in heavy industrial area.

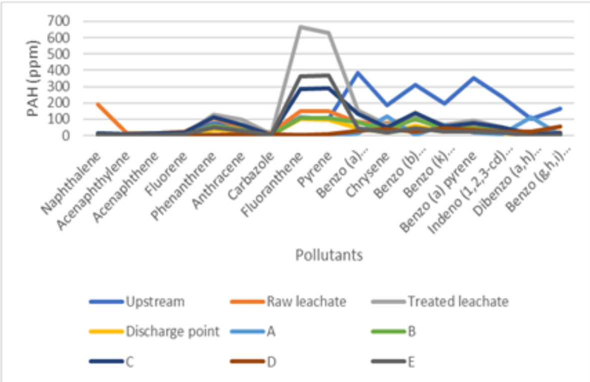

Figure 4 August- Polycyclic aromatic hydrocarbon (PAHs) ppb from Papan landfill

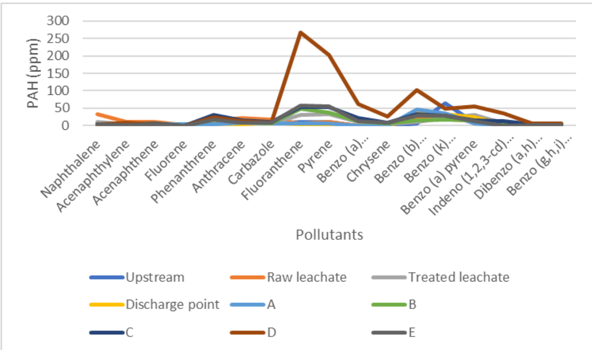

Figure 5 September- Polycyclic aromatic hydrocarbon (PAHs) ppb along Johan river

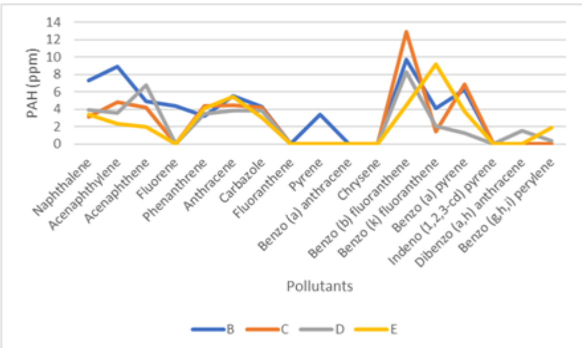

Figure 6 August- Polycyclic aromatic hydrocarbon (PAHs) ppb along Johan river

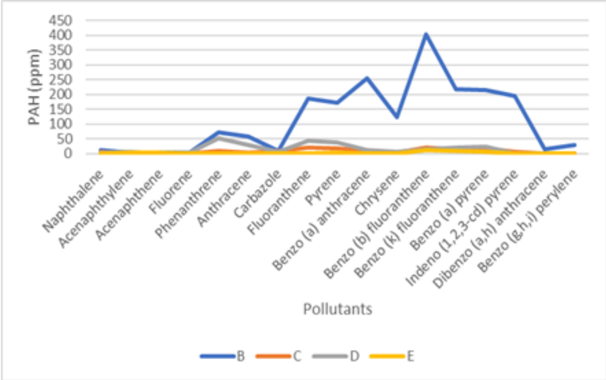

Figure 7 September- Polycyclic aromatic hydrocarbon (PAHs) ppb along Johan river

In Figure 6, pollutants shown a fluctuation trend where the highest PAH recorded was Benzo (b) fluoranthene in downstream C with concentration of 12.91 ppb. In Figure 7, it was noted that PAHs concentrations at downstream B was higher compared to the point C, D and E which includes Phenanthrene, Anthracene, Carbazole, Fluoranthene, Pyrene, Benzo (a) anthracene, Chrysene, Benzo (b) fluoranthene, Benzo (k) fluoranthene, Benzo (a) pyrene, Indeno (1,2,3-cd) pyrene, Dibenzo (a,h) anthracene and Benzo (g,h,i) perylene. This can be due to anthropogenic activities such as smoke from agricultural fires, vehicles, factory plants and other industrial sources [16]. Point B is in rural area where agricultural activities factor may take place. Thus, overall this study has proven that Johan river is unsuitable for drinking. It can cause hazard to human health such as cancer associated with the presence of PAHs.

4. Conclusion

In conclusion, this study had achieved the objectives. So far it highlighted the WQI and occurrence of micropollutants- PAHs from landfill. Parameters that are sub-indices in the WQI calculation could cause changes in the WQI values. It should be noted that the quality of a water body has been found to be very dynamic and change frequently. Thus, this study could serve as a reference for further studies on the water quality of Johan river. However, further analysis is required to determine the other micropollutants such as endocrine disruptive chemicals (EDCs) in the river water.

5. Acknowledgement

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