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Isotopic and Hydrochemistry Fingerprinting of Leachate Migration in Shallow Groundwater at Controlled and Uncontrolled Landfill Sites

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Abstract: Hydrochemical and stable isotopic monitoring were performed in two different landfill sites, which are Matang and Beriah landfills for identifying leachate migration into shallow groundwater. The data provides information and allows inferences to be made on chemical characteristics of the contaminants, migration pattern of the groundwater and the difference between controlled landfill and open dumping which depends on the groundwater quality performances for landfill. A series of comprehensive hydrochemistry data was interpreted by using a piper and stable isotope relationship diagrams. Hydrochemistry parameters were major ions (Cl⁻, SO₄²⁻, NO₃⁻, HCO₃⁻, Mg²⁺, Na⁺,Ca²⁺ and K⁺) and for the stable isotopes consist of δ²H and δ¹⁸O. The pollutans of Matang landfill shows have the tendency to migrate and disperse toward the southeast side of the landfill site and Beriah landfill site shows that the contaminants tend to migrate to the south west. This result was parallel to the isotopic composition which shows mixing between shallow groundwater with leachate. The concentrations of heavy metals are more higher in Beriah landfill compared to Matang landfill. The significant difference is due to the type of landfill itself; Matang landfill operates as a sanitary landfill and Beriah Landfill operates as an open dumping.

Key words: Groundwater • Isotope • Contamination • Hydrochemistry

INTRODUCTION

Since water covers more than 70% of the earth, water sources have always been a precious commodity for humans for a wide variety of purposes (domestic, industrial. irrigation, recreation fisheries development). However, only 1% of the earth's water is available as a source of drinking and yet our society continues to contaminate this precious resource [1]. An estimate by a World Bank report suggests that by the year 2025, about 3.25 billion people in 52 countries will live in conditions of acute water shortage [2]. This is due to the indiscriminate disposal of anthropogenic wastes and leaching of pollutants from the increase in population, urbanization and industrialization which are all a major threat to the quality of groundwater resources [3].

Besides, soils have become increasingly polluted by wastewater and agricultural chemicals (fertilizers, pesticides, herbicides) which can be easily transported in shallow groundwater [4]. Landfill holds potential contamination of readily degradable organic matter fromevery type of material, including municipal, medical, industrial, construction and radioactive waste. The detection of landfill leachate in groundwater adjacent to landfills can be complicated by the limitations of conventional monitored parameters, which are often unable to differentiate sources of pollution.

The protection of groundwater from pollution is a matter of concern to clearly characterize the groundwater quality, sources of pollution and trace the movement of pollutants from known sources [3]. The physical and chemical parameters of groundwater play a significant role in classifying and assessing water quality. Hydrochemical facies vary in groundwater aquifers due to differing lithologies [5]. Sandow [6] and Zahirin et al. [7] reported that mineral weathering, saline water intrusion,

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chemical fertilizers, organic manure, domestic sewage and the oxidation of organic carbon by nitrate are the sources of variation in hydrochemistry of groundwater.

Isotope fractionation is a geochemical tool used to identify groundwater origin, determine its age residence time and how it is affected by geo-hydrologic and meteorological factors [8]. The isotopic composition of oxygen and hydrogen are reported in terms of differences of $\delta^{18}O/\delta^{16}O$ and δ D/H ($\delta^{2}H/\delta^{1}H$) ratios relative to a standard called Standard Mean Ocean Water (SMOW) [9]. This signature serves as a natural tracer for the provenance of groundwater. It is necessary to understand the hydro-chemical characteristics and correlate with the isotopic composition for precise fingerprinting of groundwater pollution. Therefore, the main objective of this study is to evaluate the ability of the integrated stable isotope ratios of δ^2 H, δ^{18} O and δ^{13} C and hydrochemical characteristics in distinguishing the water samples analyzed and generate a potential tool in detecting leachate contamination in shallow groundwater at two different types of landfills. In doing so, a clearer picture of the situation of the water system may be identified.

Regional Setting: Both the Matang and Beriah landfill sites in this study are located in Perak state, Malaysia and field sampling of groundwater from selected boreholes, surface water from the nearest river and leachate were collected. Matang Landfill is a controlled landfill site covering a 50-acre area (Figure 1). The landfilling operations started in 1995 and since then, over 1,000,000 metric tonnes (approximately 75,000 tonnes annually) of domestic wastes have been dumped into the landfill. The site is disused mining land located in Perak. The topography in the vicinity of the landfill is generally flat and low-lying, with local elevations at the site ranging from a high of 3.3 meters above sea level to a low of 1.8 meters. The geological features of the surrounding study area have been mapped and described in detail in previous studies by Suntharalingam and Teoh [10] and Foo [11]. The type of rock consists of alluviums of the Simpang and Gula Formation, covering the eastern, northern and southern parts of the area. Alluvium of Simpang Formation is of continental origin and mainly consists of clay, silt, sand and gravel. Alluvium of Gula Formation consists of Holocene green marine clay to estuarine clay. Along the western part, the Beruas Formation, which is Quaternary alluvial deposits generally consisting of peat, is the youngest deposit in this area



Fig. 1: Perak state map and the location of Matang and Beriah Landfill

forming a belt treading from north to south. The landfill operation includes daily soil cover and leachate circulation system.

Beriah landfill is an open dumping site located in the vicinity of 5°04'N latitude and 100°35'E longitude. It covers an area of 10 acres of palm oil plantation. The site is laid on an area of mainly recent alluvium deposits which consist of silt, clays, sand and gravel. Bosch [12] reported that the peat in the Beriah area was found to overly the Holocene deposits of the Gula Formation. Beriah Landfill started its operation in 2000. Since the operation started, it has received roughly 660,000 metric tons of domestic waste. That makes the daily average of domestic waste around 200 metric tons. The climate of the area is classified as typical of the Malaysian Peninsula (equatorial) characterized by uniform temperature with daily mean minimum (30°C) and maximum (40°C) and high humidity (80 - 90%). This area has high average annual rainfall of about 1800mm.

MATERIALS AND METHODS

The sampling was conducted from November 2010 to March 2011. Groundwater from eight boreholes in Beriah and nine boreholes in Taiping were sampled by using a portable engine pump (Model Tanaka TCP 25B, maximum capacity of 110 L/min, maximum suction head of 8m and maximum delivery head of 40m). Sampling of groundwater

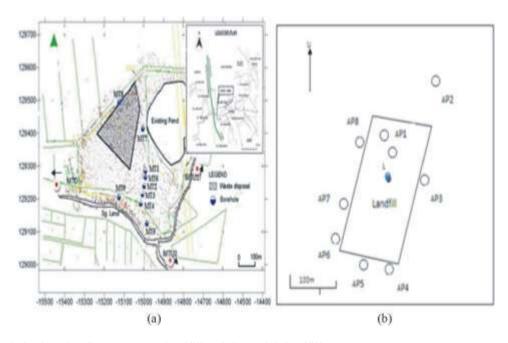


Fig. 2: Boreholes location for (a) Matang landfill and (b) Beriah landfill

was done in MT1, MT2, MT3, MT4, MT5, MT6, MT7, MT8 and MT9. Figure 2 shows the location of boreholes in the study area. Before groundwater pumping activities were carried out, water levelshad tobe taken first using the deep meter.

During sampling at the sites, the groundwater parameter was also taken using a Multiparameter probe. The parameters were measured in-situ using Multiparameter prob in the field in order to acquire representative values of ambient aquifer condition for pH, dissolved oxygen content, electric conductivity, salinity, total dissolved solids and temperature. The samples were further analyzed for major anions and cations using standard methods. Groundwater from eight boreholes was sampled in Beriah for AP1, AP2, AP3, AP4, AP5, AP6, AP7, AP8 and leachate (APL) and the sampling activies was follow the same as Matang landfill.

In hydrochemical method major ions (Anions and Cations) were measured by ion chromatography system (ICS). The ion chromatography system, ICS 1100 basic integrated IC system, was used to analyse the ground water for major ions present. This equipment uses a self regenerating suppressor and columns to detect anions such as chloride (Cl⁻), sulphate (SO₄²⁻), nitrate (NO₃⁻) and cations such as calcium (Ca²⁺), magnesium (Mg²⁺), sodium (Na⁺) and potassium (K⁺).

Hydrochemical analysis was used in interpretation of results. Major ions existing in the water samples were

analysed and discussed. The software AquaChem 5.1 was used in interpretation of major ionsand for multivariate analysis. The results of samples of different sampling time were also compared to observe the difference of chemical properties and the cause of such changes were discussed in this research. All the samples for stable isotopes were measured for δ^2 H, δ^{18} O and δ^{13} C–DIC using SerCon GEO 20–20 Continuous Flow Isotope Ratio Mass Spectrometer (CF–IRMS). For δ^2 H and δ^{18} O sampling, 100–ml HDPE bottles were fully filled with water samples to ensure that there were no air bubbles; these were then tightly capped.

Samples for $\delta^2 H$ and $\delta^{18} O$ analyses were treated in the SerCon Water Equilibration System (WES) prior to analysis through the IRMS. The δ ¹⁸O values were measured via equilibration with CO₂ at 50°C for 8 hours and δ²H values were measured via equilibration with H₂ and reaction with Platinum stick catalyst at 50°C for 1 hour. In the δ^2 H analysis, a platinum catalyst stick was used to accelerate the reaction and a gas exchange equilibrium took place between the introduced pure H₂ gas and water vapour. This gas exchange equilibrium caused the water vapour to give a signature to the introduced pure H₂ gas, which represents the isotopic composition of the water before the H₂ gas is analyzed by the IRMS. The stableisotopic composition of δ^{18} O and δ^2 H is expressed as per mil deviation (δ %) of the ratio ¹⁸O/¹⁶O or ²H/¹H with reference to the Standard Mean Ocean Water. δ‰ iscomputed by the equation

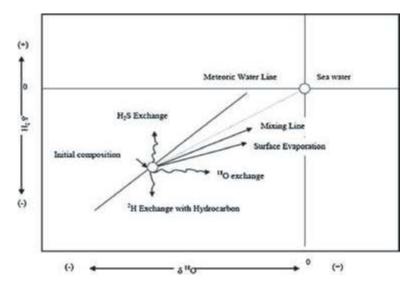


Fig. 3: Isotope exchange processes that modify the isotopic composition of meteoric water [13]

belowwhere R represents the ratio of $^{18}\text{O}/^{16}\text{O}$ or 2H/1H. The analytical errors for $\delta^{18}\text{O}$ and $\delta^{2}\text{H}$ are $\pm 0.1\%$ and $\pm 1.0\%$, respectively.

$$\delta(\%_0) = \left[\frac{R_{sample} - R_{standard}}{R_{standard}}\right] \times 1,000$$

The isotopic composition of groundwater is generally controlled by meteorological processes and is intended for the analyses of δ^{18} Oand δ^{2} H in a groundwater system. The result of the isotope exchange process using the δ^{18} O and δ^{2} H diagram is a deviation from the meteoric water line along a line with a lower slope, which depends onthe relative humidity. However, some extreme geological environments exist where the reaction between groundwater and the aquifer matrix or subsurface gases could change the meteoric signature of the water. Some typical processes are shown in Figure 3.

RESULTS AND DISCUSSION

Hydrochemistry: Results from hydro-chemical analysis can be plotted as a Piper diagram in the form of a graph and calculation using AQUACHEM. The piper diagram consists of 3 triangles with each divided into different quadrants. The quadrant in which the plot of the sample falls in is classified according to the ion groups stated. The Piper diagram is a type of triliner diagram broadly used in hydrogeology as it illustrates the hydrochemical

characteristics of the groundwater by representing the concentration of anions and cations in separate triangular diagrams [14]. The diagram has been classified into four hydrochemical facies based on the dominance of different cations and anions.

From Figure 4 (a) and Table 1, it is observed that samples from MT4, MT6, MT5-2, upstream and downstream falls under Type I while MT1, MT2, MT3, mines pond water and leachate fall into Type II. For Type III and IV, the samples that fall under it were MT8, MT9 and MT7, MT5-1 respectively. It can also be seen that the amount of Alkalis (Na $^+$ -K $^+$) exceeded the alkaline earth (Ca $^{2+}$ – Mg $^{2+}$) and weak acids (CO $_3^{2-}$ - HCO $_3^{-}$) exceed strong acids (Cl $^-$ - SO $_4^{2-}$).

Figure 4 (b) belowshows the Piper Diagram for March sampling. From the diagram, MT4, MT6, MT5-2, upstream and downstream fall under Type I, the same as the first sampling. MT1, MT2, MT3, mines pond water and leachate still remainunder Type II but MT7 and MT5-1 changed into Type III and the rest still the same as the January sampling. The amount of Alkalis (Na+K+) exceeded the alkaline earth (Ca²⁺ – Mg²⁺) and weak acids (CO₃² - HCO₃⁻) exceed strong acids (Cl⁻ - SO₄²-). Based on the results for both samplings, it can be said that water samples from upstream and downstream are naturally undisturbed since it is in Type I facies. Natural groundwater generally has higher concentration of Ca²⁺ and Mg²⁺ ions and bicarbonate ions as well. The natural groundwater is alkaline due to prolonged exposure to rock structures that contains high amounts of Ca2+ and Mg²⁺ ions.

Table 1: Water Type on the Basis of Piper Diagram for Matang Landfill Site

Facies	Water Type		January 2011 sampling	March 2011 sampling		
1	Ca ²⁺ - Mg ²⁺ - HCO ₃ -	Type I	MT4, MT6, MT5-2, Upstream, Downstream	MT4, MT6, MT5-2, Upstream, Downstream		
2	$Na^+ - K^+ - HCO_3^-$	Type II	MT1, MT2, MT3, Mines Pond water, Leachate	MT1, MT2, MT3, Mines Pond water, Leachate		
3	$Na^+ - K^+ - Cl^ SO_4^{2-}$	Type III	MT8, MT9	MT5-1, MT7, MT8, MT9		
4	$Ca^{2+} - Mg^{2+} - Cl^{-} - SO_4^{2-}$	Type IV	MT5, MT7	-		

Table 2: Water Type on the Basis of Piper Diagram for Beriah landfill

Facies	Water Type		January 2011 sampling	March 2011 sampling
1	Ca ²⁺ - Mg ²⁺ - HCO ₃ -	Type I	-	AP3
2	$Na^+ - K^+ - HCO_3^-$	Type II	-	AP2
3	$Na^+ - K^+$ - Cl^- - SO_4^{2-}	Type III	AP1, AP2, AP3, AP5, AP6, AP7, AP8 and APL	AP4 and AP1
4	$Ca^{2+} - Mg^{2+} - Cl^{-} - SO_{4}^{\ 2-}$	Type IV	AP4	APL, AP5, AP6, AP7 and AP8

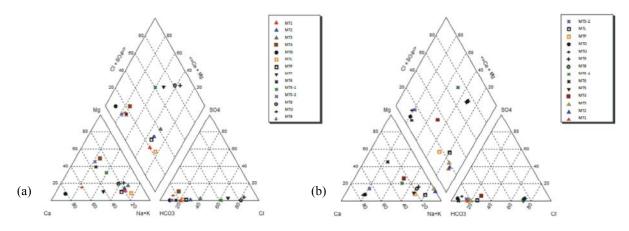


Fig. 4: Matang landfill piper diagram for (a) January 2011 and (b) March 2011 samplings.

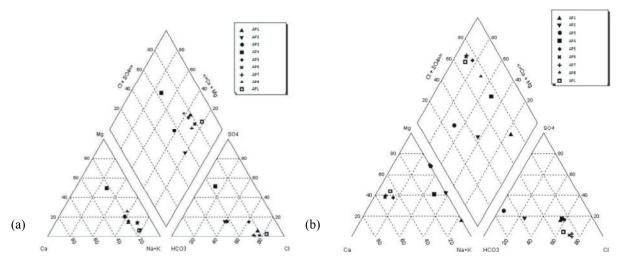


Fig. 5: Beriah landfill piper diagram for (a) January 2011 and (b) March 2011 samplings

Figures 5 (a) and (b) show the Piper Diagram for Beriah Landfill in excess the water quality including leachate at Beriah landfill site. The type of facies for the January and March samples are shown in Table 2 and there are differences in water type for both samplings. The January sampling shows all the samples were in Type III that is $Na^+ - K^+ - Cl^- - SO_4^{2-}$. Compared to the January sampling, most of the March samples are in Type IV

Ca²⁺ – Mg²⁺ – Cl⁻ – SO₄²⁻. The similar water types of AP5, AP6, AP7, AP7 and AP8 with leachate for both samplings described the leachate influence to the shallow groundwater in Beriah landfill. The types of leachate itself differ between both sites where for Matang, leachate falls in TypeII whereas Berriah landfill leachate was in Type III and Type IV. The difference could be due to the different chemical reactions in different landfill types and rainfall intensity where Matang has higher a average annual rainfall of (400 mm) compared to Beriah (1800 mm) that influenced the characteristics of the samples. The amount of Alkalis (Na⁺-K⁺) exceeded the alkaline earth (Ca²⁺ – Mg²⁺) and weak acids (CO₃²⁻ - HCO₃⁻) exceed strong acids (Cl⁻ - SO₄²⁻).

Heavy Metal Distribution: Heavy metals have been into the environment due industrialization and have created major global concern [15]. A previous study identified heavy metals such as zinc, arsenic, lead and cadmium in landfills leachate [16]. Leachate produced from the generation of solid wastes can infiltrate the soils and groundwater. Based on the topography and colloidal borescope system measurement, the regional groundwater flow direction is dominantly towards the south and southeast of the study area [17]. Groundwater typically moves in parallel paths, or layers. Since groundwater movement is slow, it doesn't create enough turbulence to mix the way surface waters mix when a river or stream empties into another waterbody. That is, layers of groundwater remain relatively intact. Diffusion is a very slow process, but over a period of years, it can create significant pollution problems around the landfills.

Table 3 shows the concentration of heavy metal in shallow groundwater for both landfill sites. Heavy metals

that were analysed in this research were Iron (Fe), Nickel (Ni), Chromium IV (Cr⁶⁺) and Copper (Cu). The distribution of Fe at both site samples shows different values. The highest Fe content of 10.5 mg/l is measured in groundwater sample, AP7 for Beriah landfill and 6.7mg/L in leachate sample for Matang landfill. The high level of Fe in leachate indicates that the Fe and steel scrap dumped into the landfill may be due to the iron and tin based waste in landfill site [18]. The high value of Iron in groundwater sample is due to the close proximity to the landfill. Studied by Suman [19], the leachate is generally strongly reducing liquid formed under methanogenic conditions and upon coming into contact with aquifer materials, has the ability to reduce sorbed heavy metals in the aquifer matrix and the most important reaction of Fe and Mn to more soluble species. Results from Matang Landfill show the concentration of Cu is relatively lower, less than 1mg/L except for MT6 which is 1.26mg/L. The highest concentration of Cu is found in Beriah Landfill groundwater sampleMT2, which is 4.5mg/L. The highest value of Ni is found in MT3 (Beriah Landfill), 4.8 mg/L while the lowest value of Ni is found in MT5 (Matang landfill), 1.21 mg/L. The Ni concentrations are a result of municipal origin, mainly from discarded waste items such as refused batteries, paint products, metallic items and fluorescents lamps [20]. Cr concentrations in Matang Landfill are low which less than 1 mg/L for all samples. However, for Beriah Landfill, the highest concentration is found in MT2, which is 6.7 mg/L. Concentration of Cr reveals the presence of wood preservatives and paint products in the waste [20]. Figure 6 shows iron distribution in both Beriah and Matang landfill. Figure 6 (a) shows the highest iron concentration is at AP7 with 10.5 mg/L, whereas MT1, MT2 and MT6 give the highest reading for Matang landfill within 4 - 5 mg/L.

Table 3: Mean heavy metal concentartion for Matang and Beriah landfill sites

Samples		Fe (mg/L)		Cu (mg/L)	Cu (mg/L)		Ni (mg/L)		Cr (mg/L)	
Matang	Beriah	Matang	Beriah	Matang	Beriah	Matang	Beriah	Matang	Beriah	
MT1	AP1	5	2.5	0.55	1.75	0.73	1.1	0.05	1.15	
MT2	AP2	4.3	0.5	0.41	4.5	0.63	1.85	0.04	6.7	
MT3	AP3	1.76	2	0.38	0	0.55	4.8	0.04	0	
MT4	AP4	0.9	3.25	0.31	0.25	0.51	1.35	0.04	0.5	
MT5	AP5	0.06	6.75	0.07	0.5	0.1	0.4	0.02	0	
MT6	AP6	2.65	7.5	1.25	2.25	0.6	3.4	0.04	3.35	
MT7	AP7	0.32	10.5	0.27	0	0.42	3.35	0.03	4	
MT8	AP8	0.04	0.5	0.12	0.5	0.48	1.88	0.01	1.45	
MT9		0.21		0.12		0.24		0.02	1	
LEACHATE	LEACHATE	6.7	3	0.65	0.5	1.21	3.5	0.08	3.05	

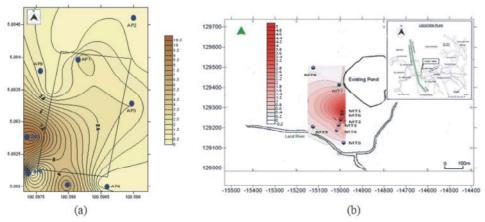


Fig. 6: Mean distribution pattern of Iron concentration at (a) Beriah landfill and (b) Matang landfill

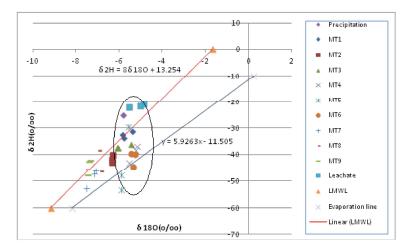


Fig. 7: Plot of δ²H vsδ¹⁸O (‰) for all the groundwater samples fromMatang landfill site.

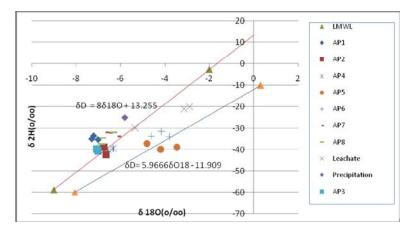


Fig. 8: Plot of $\delta^2 H$ vs $\delta^{18}O$ (‰) for all the groundwater samples from Beriah landfill site

Stable Isotopes: A previous study conducted by Tadza *et al.* [21] reported that the values of leachate for $\delta^2 H$ from Matang landfill site ranged from -9.9 to -28.8%. These were comparable to the $\delta^2 H$ values for

all the samplings of this research -in the range of -19.22 $\%_{\infty}$ to -22.02 $\%_{\infty}$. The highly enriched $\delta^2 H$ in H_2O of leachate was due to extensive methanogenesis in the landfill [22, 23].

Figure 3 shows that anaerobic activities will change hydrogen to H₂S and CH₄. Therefore the remaining hydrogen in aqueous medium was enriched in δ^2 H. δ^2 H leachate enrichment may also occur due to isotopic exchange to H₂S which happens in the reduction process of SO_4 [23]. The groundwater isotopic value for δ^2H and δ^{18} O ranged from -29.45°/ $_{00}$ to 53.40°/ $_{00}$ and -5.17°/ $_{00}$ to - $8.34^{\circ}/_{\circ o}$. Highly enriched values of δ^2 H for MT1, MT2, MT3, MT4 and MT6, as seen in Figure 7, indicated leachate influenced the groundwater characteristics. Groundwater had mixed with leachate and resulted in the shifting of δ^2 H value. This shows that leachate from the landfill flows towards the southeast of the landfill. The leachate for Beriah presents (Figure 8) the enrichment of $\delta^2 H$ that is in the range of -25.15°/ $_{\infty}$ to -23.11°/ $_{\infty}$ and this characteristics is most similar in AP6 and AP5 of all the groundwater. This shows the significant correlation of leachate and groundwater samples which are AP6 and AP5 (see in circle in Figure 8).

The leachate also shows the enrichment of δ^2H due to extensive methanogenesis but the value of $\delta^{18}O$ is significantly high and is located near to the evaporation line[21]. Evaporation may occur due to continuous evaporation in the leachate treatment ponds and leachate runoff. At a higher temperature or in a geothermal process, lighter oxygen-16 was easier to vapor and left the leachate with heavier isotope that is $\delta^{18}O$. This is also called $\delta^{18}O$ shift, where it becomes enriched and differentiated to other samples' isotopic characteristics. Figure 8 shows AP5 and AP6 located in the evaporation line and this was because of the leachate migration and influences the groundwater characteristics on both points.

The difference between the leachate isotopic composition of Beriah and Matang landfill are caused by the landfill type where Matang landfill is a controlled landfill where its leachate does not influences by evaporation and more to anaerobic condition due to the daily soil cover used and existing leachate circulation system. Beriah landfill is an uncontrolled landfill with no daily soil covering. As a result, leachate is exposed to evaporation with no circulation system, meaning that the leachate flows through open runoff. Therefore, the distinct isotopic characteristics of the leachate are highly affected by evaporation.

CONCLUSION

The use of stable isotope technique in this study provides a result with distinct isotopic signature for the groundwater samples studied for both landfills which have highly enriched values for δ ²H and δ ¹⁸O shift indicates, leachate influence in MT1, MT3, MT4 and MT6 for Matang and AP5 and AP6 for Beriah landfill. Additionally, the isotope technique presents a clearer picture of the situation on leachate contamination although it looks simple and straightforward. The hydrochemistry technique shows similar results in Beriah landfill where AP5 and AP6 with additional AP7 give significant water type with leachate for both samplings describe the leachate influence to the shallow groundwater. Isotope analysis coupled and assisted with hydrochemistry technique and knowledge on the area local geology are reliable in assessing the contaminantion in shallow groundwater sample due to landfill leachate.

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