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Application of Deuterium and Oxygen-18 to Trace Leachate Movement in Bantar Gebang Sanitary Landfill

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ABSTRACT

Bantar Gebang landfill was constructed in 1986 with total area of 108 ha and approximately 6000 ton/day solid waste is disposed to this landfill. Mostly, the people living surrounding landfill get afraid of impact of the hazardous chemicals produced by waste disposal to their health. The purpose of this investigation was to study the migration of leachate to Cibitung River water and shallow groundwaters near to the river. It is possible to be done because chemical contents and isotopic characteristics of municipal landfill leachate are unique, relative to aqueous media in the most natural environments. Laser absorption method developed by the LGR (Los Gatos Research) was used to measure absolute abundances of ^{2}HHO, HH^{18}O and HHO in a number of water samples. In-situ measurements were also conducted as an additional parameter besides their isotopes. The δ^{2}H of the H_2O in landfill leachate was significantly enriched, with values of - 22.6 ‰ to + 4.3 ‰. This deuterium enrichment was undoubtedly due to the extensive production of microbial methane within the limited reservoir of the landfill. However, the enriched deuterium value in leachate was not detected in the river which still had depleted values. It was probably caused by the amount of natural water in the river was comparatively large, with respect to limited leachate discarded to the river. The electrical conductivity of the leachate was higher (3200 to 7600 μS) and the decreasing values were still monitored in the river to approximately 12 km after streaming the landfills. The effect of the high electrical conductivity and enriched deuterium of leachate was not clearly indicated in the groundwater samples which still represented the local precipitation recharge, except a monitoring well located in Bantar Gebang landfill area which has an indication of leachate contamination.

INTRODUCTION

Bantar Gebang landfill is located in Bantar Gebang district, Bekasi regency - West Java Province. The rate of urbanization in Bantar Gebang increase rapidly that could be as an impact of higher industry growths in Bekasi which is one of supporting areas for Jakarta. The higher population in Bantar Gebang come up as an impact of economic crisis that taken place since the year 1997. Similar to other urban areas, some irregular settlements that are illegally built in Bantar Gebang landfill area also increase significantly [1].

Bantar Gebang landfill consists of 5 zones with total area of 108 ha and covers three villages, those are Ciketing Udik, Sumur Batu and Cikiwul. This landfill was constructed by Jakarta municipal in 1986. The purpose of constructing this landfill is to dispose the solid waste especially for organic materials produced by Jakarta people [1]. Actually, not only organic material wastes that are disposed to Bantar Gebang landfill but also non organic waste such as plastics and metals are disposed there. Waste composition produced in Jakarta is 65% and 35% for organic materials and non organic materials, respectively. It is predicted that the total waste deposition produced by 10.931.207 people living in Jakarta increase to 29.624 m^3/day in 2010. It means that about 6000 ton/day of solid waste is disposed to Bantar Gebang landfill [2].

Bantar Gebang landfill is constructed by applying a sanitary landfill system in which solid waste is filled in digged area, compacted and covered with soils. These activities are repeated for the next disposal. Bantar Gebang sanitary landfill has been improved with a system for leachate

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collection pipes, leachate treatment ponds, gas venting pipes and groundwater monitoring wells. The bottom of this landfill has been also protected by lining system made of synthetic material [3]. However, the local people generally still wonder to the quality of their environment whether their groundwater has been contaminated by the leachate.

The other secondary pollutions generated from landfill site are gases, heavy metals, offensive odor and pets or flies. The main gases are methane, ammonia, hydrogen sulfide and carbon dioxide generated from waste itself or through waste decomposition process [3]. Those gases and other hazardous chemicals are dissolved in leachate or released to the air that can cause groundwater contamination and air pollution. Finally, this result can decrease the quality of local people health.

According to the some investigations, the content of some chemicals (Fe, NO_2^-, NO_3^-, Mn and Pb), COD, BOD and pH in Bantar Gebang groundwater as a source for drinking water has been over limit to the value for drinking water quality standard related to the regulation of Health Minister No.416/MENKES/Per/IX/1990 and it is concluded that the quality of their groundwater is very worst [4]. The local people also suffer some diseases such as teeth carries, skin infection, anemia, acute respiratory infections and dysentery that might be an effect of landfill activity [1].

To support chemical data for groundwater studies, natural isotope technique offers finger printing tools that can not be solved by chemical parameter solely. All water have "finger prints" of naturally occurring isotopes that provide information about their origin. Among the most powerful and cost effective fingerprinting tools are the ratios of the stable isotopes of deuterium to hydrogen ($^2H/^1H$) and oxygen-18 to oxygen-16 ($^{18}O/^{16}O$). The ratio of both isotopes in precipitation varies according to the elevation, and its values are preserved once the precipitation infiltrates to the aquifer. Thus, scientists can use to determine the recharge location for a groundwater [5,6].

Fractionation effect in groundwater can change $^2H/^1H$ and $^{18}O/^{16}O$ isotopic ratios. For example, evaporation of water causes an increasing in both isotope ratios, on the other hand, dilution by precipitation results a decreasing in both isotope ratios. In the case of landfill sites, $^2H/^1H$ ratios from leachate can take place a shift to positive range due to the huge methane production. It means that $^2H/^1H$ ratios from leach water fall outside of the ranges associated with natural groundwater. The anomaly values of $^2H/^1H$ ratios in groundwater surrounding landfill sites can be used as an indication of leachate migration [5].

The purpose of this investigation is to understand the natural process involving isotope of deuterium (δD) and oxygen-18 ($\delta^{18}O$) in leachate water from Bantar Gebang landfill. Then, these data could be utilized as a data base if chemical and biological reactions of constituents dissolved in leachate significantly effect to the isotopic composition of Cibitung river water and their shallow groundwater. It is necessary because leachate treated from Bantar Gebang landfill is streamed to Cibitung River through the some drainage ditches. In addition, the groundwater closed to the river is also investigated to assure a migration effect of leachate water.

EXPERIMENTAL METHODS

Materials

The materials used were LGR working standard #3 ($\delta^{18}O$ = -11.54 ± 0.1 ‰ vs SMOW ; δD = -79.0 ± 1.0 ‰ vs SMOW), LGR working standard #4 ($\delta^{18}O$ = -7.14 ± 0.1 ‰ vs SMOW ; δD = -43.6 ± 1.0 ‰ vs SMOW), LGR working standard #5 ($\delta^{18}O$ = -2.96 ± 0.1 ‰ vs SMOW; δD = -9.8 ± 1.0 ‰ vs SMOW), distilled water and drierite: anhydrous $CaSO_4$.

Equipments

The equipments used were Los Gatos Research (LGR) DT-100 Liquid Water Stable Isotope Analyzer, vacum pump, glass column, CTC LC-PAL liquid auto sampler, Hamilton micro liter syringe, Polytetraflouroethylene (PTFE) septum, PTFE transfer line; 1,5 ml glass vials with cap and septa from PTFE, auto sampler tray, 1 ml automatic pipette, 1 ml disposable pipette tips, a set of computer and printer, Anotop 10 membrane filter 0.2 µm; 5 ml and 1.25 µL syringe, 20 ml glass vials with screw caps, water sampler, Magellan SporTrak Global Positioning System and conductivity meter.

Sampling methods

Leachate waters were collected either from leachate treatment ponds or leachate run off from solid waste dump in Bantar Gebang landfill and Sumur Batu landfill during March to April 2010. Cibitung River water samples were collected in the beneath the air-surface interface with distance about 3 km from sites whereas groundwater samples were collected as closed as possible to Cibitung River sites. The 20 ml glass bottles were fully filled with water. The cap was placed on the top of bottle while still out of contact with air and sealed tightly.

Water samples were always kept such that they were not in contact with air until opened and ready for analysis in the laboratory of Hydrology – Center for the Application of Isotopes and Radiation Technology, National Nuclear Energy Agency. Conductivity of each sample was measured using in-situ technique.

Analysis of δ D and $\delta^{18}O$

In the laboratory, to measure stable isotope ratios of hydrogen and oxygen were used LGR DT-100 Liquid Water Stable Isotope Analyzer which is connected to CTC LC-PAL liquid auto sampler by PTEE transfer line as shown in Fig. 1. Briefly, 1 ml waters samples (filtered if it is cloudy or contains of sediments) and three kinds of working standards were dropped into 1.5 ml auto sampler glass vials and closed with septum caps. Those standards and samples were arranged in a certain order in auto sampler tray such that every three standards were followed by five samples. Meanwhile, both equipments were turned on at least three hours to stabilize them and also to heat injection port auto sampler to 80^{o}C.

As much as 0.75 μL water sample was injected to injection port auto sampler at CTC LC-PAL through 1 m PTEE transfer line. In which, water sample was evaporated and then the vapor traveled through transfer line into the pre-evacuated mirrored chamber for analysis. A laser beam was directed to through vapor sample and the mole fraction of a gas was determined from measured absorption using Beer' Law as a conventional spectroscopy. To reach adequate precision in measured isotopic ratio, the LGR instrument utilizes an Off-Axis Integrated Cavity Spectroscopy (off axis ICOS) approach which can generate optical path length of about 2500 m in a 25 cm cell. The molecular concentrations of ^{2}HHO, HH^{18}O and HHO were calculated by measuring the amount of absorbance at a wavelength of 1390 nm. Each sample was measured six times to obtain good reproducibility values [7,8].

Molecular concentrations were converted into atomic ratios, $^{2}H/^{1}H$ and $^{18}O/^{16}O$ and then the delta (δ) values were calculated according to Vienna Standard Mean Ocean Water (VSMOW) as the follows [7]:

$$\delta = \frac{R_{measured} - R_{VSMOW}}{R_{VSMOW}}$$

R is isotopic ratio of $^{2}H/^{1}H$ or $^{18}O/^{16}O$.

Fig. 1. LGR DLT-100 liquid – water stable isotope instrument and CTC LC-PAL liquid auto sampler.

RESULTS AND DISCUSSION

Previously, the investigation employing other isotope such as oxygen-18 and sulphur-34 from dissolved sulphates as an addition for chemical parameters in Bantar Gebang shallow groundwater had been carried out. The results suggested that dissolved sulphate of shallow groundwater near to landfill area might have been influenced by leachate water [9]. Syafalni [10] reported that domestic shallow wells in Bantar Gebang are becoming increasingly polluted by nitrate in the regions which have been distributed more than 1 km from landfill site. Based on oxygen-18 and deuterium, it was concluded that evaporation process had taken place in shallow groundwater surrounding of this landfill.

This investigation was stressed to Cibitung River sites as long as 12 km after streaming to Bantar Gebang landfill. It is important because this river is utilized to discard leachate water which had been treated from leachate collection ponds either from Bantar Gebang or Sumur Batu landfill. The groundwater sites were collected as closed as possible to this river in order to know whether leachate dissolved in river water had an effect to their shallow groundwaters. Mostly, the influence of surface water, such as river or seawater, which might have been contaminated toward groundwater system as a main source of drinking water supply must be eliminated. The term of shallow groundwater referred to the unconfined aquifer system with the depth scale around 0-40 m [11]. Sample types and their coordinates for Cibitung River, shallow groundwater and leachate water sites in surrounding of Bantar Gebang landfill were shown at Table 1, whereas their locations were mapped in Fig. 2. The results including electrical conductivity, O-18 and deuterium isotopes contained in H_2O compounds were presented at Table 2.

Table 1. Sampling locations in Bantar Gebang landfill area.

No	Sample ID	Sample type	Coordinate Longitude - Latitude
1	R-1	Cibitung River	06°21'51" – 106°58'47.2"
2	R-2	Cibitung River	06°22'05.2" – 106°59'09.9"
3	R-3	Cibitung River	06°21'17.4" – 106°58'58.3"
4	R-4	Cibitung River	06°20'43.9" – 106°59'55.9"
5	R-5	Cibitung River	06°20'34.3" – 107°00'09.8"
6	R-6	Cibitung River	06°19'55" – 107°00'16.2"
7	R-7	Cibitung River	06°'19'15" – 107°01'01.9"
8	R-8	Cibitung River	06°18'31.9" – 107°01'31.7"
9	R-9	Cibitung River	06°17'26.1" – 107°01'38.5"
10	R-10	Cibitung River	06°16'19.4" – 107°02'00.5"
11	L-4a	Leachate-3 (inlet - surface) -Bantar Gebang	06°21'12" – 106°59'50.1"
12	L-4b	Leachate-3 (pond-1.5 m depth)-Bantar Gebang	06°21'12" – 106°59'50.1"
13	L-4c	Leachate-3 (outlet - surface)-Bantar Gebang	06°21'12" – 106°59'50.1"
14	L-4d	Leachate run off - Bantar Gebang	06°20'50.1" – 106°59'37.4"
15	L-4e	Leachate run off - Bantar Gebang	06°20'43.8" – 106°59'41.5"
16	L-5	Leachate (pond - surface) - Sumur Batu	06°20'35.1" – 107°00'10.2"
17	G-2	Groundwater – 30 m depth	06°22'03.8" – 106°59'17.6"
18	G-3	Groundwater – 8 m depth	06°21'15.3" – 106°58'56.5"
19	G-4	Groundwater – 6 m depth	06°21'12" – 106°59'50.1"
20	G-5a	Groundwater – 10 m depth	06°20'35.1" – 107°00'11.4"
21	G-5b	Groundwater – 40 m depth	06°20'41.0" – 107°00'15.8"
22	G-6	Groundwater – 5 m depth	06°19'48.7" – 107°00'16.2"
23	G-7	Groundwater – 10 m depth	06°'19"7.8" – 107°00'45.9"
24	G-8	Groundwater – 10 m depth	06°18'19.7" – 107°01'30.1"
25	G-9	Groundwater – 40 m depth	06°17'48.4" – 107°01'45.9"
26	G-10	Groundwater – 8 m depth	06°16'18.9" – 107°02'13.5"

Wide range of conductivity values of rive samples, 62.1 µS to 689 µS, was observed wherea conductivity values of groundwater samples were similar with the mean value of 211.4 µS, except G-4 sample having a much higher value i.e 1018 µS. G-4 is located in landfill site of Bantar Gebang and constructed as a monitoring groundwater well for dumping area in zone-3 (approximately 3 m from leachate treatment pond). Thus, the chemicals from leachate water would be easier to move to G-4.

In contrast, conductivity of leachate water samples was distributed at a very high level from 3200 µS to 7600 µS, except leachate water from Sumur Batu landfill having a lower level. Sumur Batu landfill is especially utilized for Bekasi regency waste dump constructed in 2002 with smaller capacity (10 ha) [12] than Bantar Gebang landfill (108 ha). Thus, the rate of organic waste decomposition which produced HCO_3^-, H_2S, NH_3 gas; dissolution cations and anions; redox reactions and other reactions was not as extremely high as in Bantar Gebang landfill. Figure 3 also showed the trend of decreasing conductivity value of Cibitung River after streaming both landfill sites. However, conductivity value at the distance about 12 km from landfill site was still detected at a higher value than those of three sites before the river streamed to landfill.

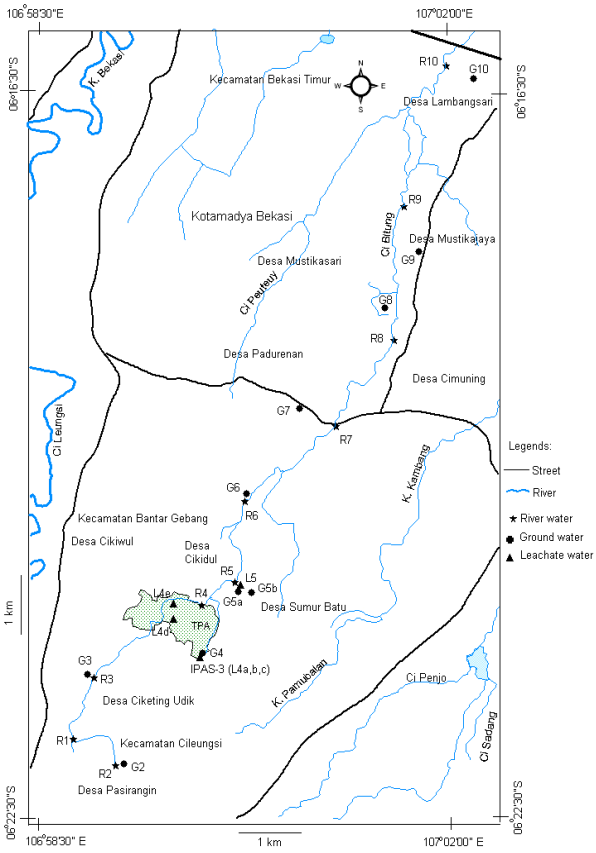

Fig. 2. Sampling sites for Cibitung River, leachate and groundwater.

The leachate directly running off had a higher conductivity level than those in leachate after treating in ponds. It might be due to higher inorganic ion contents such as Na^+, K^+, Ca^{2+}, Mg^{2+}, Fe^{2+}, Mn^{2+}, Cl^-, SO_4^{2-}, and HCO_3^-. The origin of the major cations in leachate was possibly derived from the chemical sources in refuse, alteration of silicate minerals (such as pyroxene, epidote bearing ferro-magnesium silicates) or desorption from clay. The later might be most important source of those

major cations because clay was daily used as cover material of the trash. In this case, ionic exchange reaction occurred between major cations from clay and large amount of ammonium generated from refuse [13].

Table 2. Isotopic data for leachate water, Cibitung River and groundwater in Bantar Gebang site

No.	Sample ID	Electrical conductivity (µS)	$\delta^{18}O_{SMOW}$ (‰)	$\delta^{2}H_{SMOW}$ (‰)
1	R-1	62.4	-6.54	-39.2
2	R-2	235	-6.65	-40.3
3	R-3	73.2	-6.4	-42.2
4	R-4	615.6	-6.62	-41.1
5	R-5	689	-6.2	-40.3
6	R-6	654.9	-7.56	-42.9
7	R-7	642	-6.72	-38.9
8	R-8	568	-6.34	-40.4
9	R-9	582	-6.7	-39.2
10	R-10	307	-6.25	-38.9
11	L-4a	4150	-5.19	-22.6
12	L-4b	3430	-4.21	-23.4
13	L-4c	3280	-4.22	-22.8
14	L-4d	7590	-5.58	+2
15	L-4e	7610	-5.55	+4.3
16	L-5	843	-4.45	-25.4
17	G-2	122	-6.83	-34.8
18	G-3	210	-7.1	-42.8
19	G-4	1018	-5.77	-31
20	G-5a	168.7	-6.05	-36.7
21	G-5b	229	-6.81	-38.2
22	G-6	186.9	-6.28	-36.3
23	G-7	252	-5.92	-38.8
24	G-8	289	-6.9	-37.4
25	G-9	220	-7.33	-40.9
26	G-10	225	-6.55	-37.7

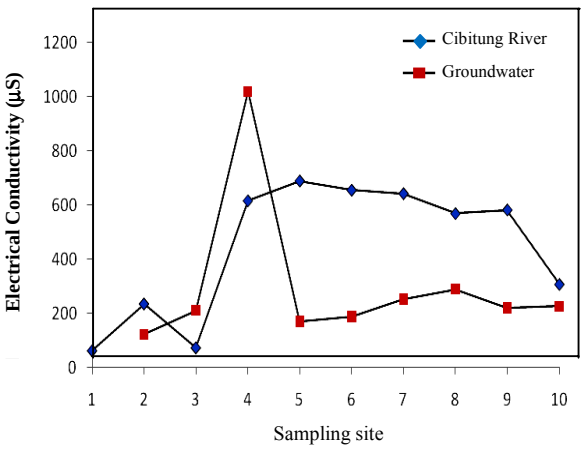

Fig. 3. Conductivity values for Cibitung River and groundwater.

As mentioned above that deuterium and oxygen-18 were made to determine if constituents of dissolved leachate had a significant effect to the isotopic composition of groundwater and river water. Figure 4 showed the relationship of $\delta^{2}H$ versus $\delta^{18}O$ data for local meteoric water [14] and three kinds of samples observed. Mostly, those natural water samples observed followed the trend of local meteoric water line as an initial source of groundwater, except leachate water samples. The values of $\delta^{18}O$ and $\delta^{2}H$ for Cibitung River ranged from -7.56 ‰ to -6.20 ‰ with mean value of -6.60 ‰ and -42.9 ‰ to -38.9 ‰ with mean value of -40.36 ‰, respectively. Their shallow groundwater closed to Cibitung River gave rather more enriched values of -7.33 ‰ to -5.77 ‰ for $\delta^{18}O$ with mean value (except G4) of -6.64 ‰ and -42.8 ‰ to -31 ‰ for $\delta^{2}H$ with mean value (except G4) of -38.18 ‰. The more depleted deuterium values in Cibitung River might be due to a mixing process among water sources which recharged at different elevations. Local precipitation at approximately elevation of 140 m had $\delta^{18}O$ of -6.21 ‰ and $\delta^{2}H$ of -34 ‰ whereas the region at higher elevation about 1020 m had $\delta^{18}O$ of -7.98 ‰ and $\delta^{2}H$ of -49.4 ‰ [14]. From deuterium data of local precipitation, the shallow groundwater samples located near to Cibitung River were probably influenced by local precipitation. However, oxygen isotope values for the river water and groundwater in this investigation did not differ significantly. It suggested that there were any water interactions between Cibitung River and shallow groundwater located closed to the river or they had same water origin (these values were similar to $\delta^{18}O$ from local precipitation). Site of G-4 is located more than 1 km from the river and surrounded by hilly refuse, it gave anomalous natural isotope values. The more enriched values of $\delta^{2}H$ and $\delta^{18}O$ of G-4 than those of other groundwater samples indicated that it was influenced by leachate migration which also had more enriched values for both isotopes.

Interesting values appeared at leachate samples collected from two active landfill sites having unusual heavy isotope values, especially for ^{2}H. The $\delta^{2}H$ and $\delta^{18}O$ of leachate ranged from -22.6 ‰ to +4.3 ‰ and -5.58 ‰ to -4.21 ‰, respectively. The highly enriched values for deuterium in leachate relative to the average $\delta^{2}H$ of local precipitation were also observed in leachates from three active landfills in Illinois – USA which had range values of deuterium from -60.0 ‰ to +14.5 ‰ [13]. The $\delta^{2}H$ value of leachate collected from four landfills in New Zealand ranged from -47 ‰ to -4 ‰ [15]. It was undoubtedly that the enriched values of deuterium in leachate were a result of the very active formation of microbial methane, producing methane gas, within a closed system over time. In the case municipal landfills or

the surrounding leachate, a significant amount of hydrogen incorporated in the CH_4 (25% to 100%) came from the surrounding water during methanogenesis. The microbes preferentially utilized the isotopically light hydrogen when producing CH_4. Therefore, the remaining hydrogen in aqueous medium was enriched in deuterium. In the relatively closed environment of landfills where the generation of microbial methane was much larger than normal reservoir of water, it was possible to observe this deuterium enrichment. Besides the effect of methanogenesis, it was possible that some deuterium enrichments observed in landfill leachate were due to isotopic exchange with H_2S, which would be generated during SO_4 reduction in landfills. However, methanogenesis process was probably major effect producing deuterium enrichment in leachate relative to isotopic exchange reaction [5,13].

The $\delta^{18}O$ of leachate samples in this investigation slightly shifted to more enriched values than that of local precipitation value. It might be due to high evaporation effect which carried out continuously in open leachate treatment ponds and leachate run off. In higher temperature, a lighter isotope of oxygen-16 in H_2O compounds in liquid phase (in this case is leachate) was easier to move to vapor phase. Therefore, the remaining of liquid phase would be enriched in heavier isotope of oxygen-18.

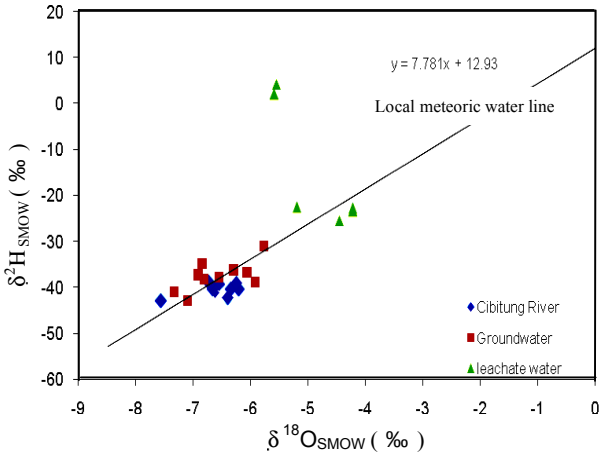

Fig. 4. Deuterium vs Oxygen-18 values for Cibitung River, groundwater and leachate.

The highly enriched deuterium values of all leachate samples (including leachate ditches) in this investigation did not influenced significantly to those of Cibitung River water samples. It could be explained that the rate of river water flow originating from higher elevations (containing more depleted deuterium values) was much faster than the rate of mixing process with leachate flow. Moreover, the amount of natural water in the river was comparatively large, with respect to limited leachate discarded to the river. Additionally, the rainfall during sampling period in Bogor area, as an upstream of Cibitung River, was still high in scale from 300 mm to 400 mm [16]. Based on the different δ^2H data between leachate and river water, the effect of leachate flow toward Cibitung River could not be observed clearly. However, chemical data represented by higher conductivity value of the river after streaming the landfill sites indicated that Cibitung River was contaminated by leachate.

CONCLUSION

Leachate samples had significant enriched values of δ^2H in the H_2O ranging from -22.6‰ to +4.3‰ which were due to extensive production of microbial methane within the limited reservoir of the landfill. However, the enriched deuterium values in leachate could not be traced in the Cibitung River which still had depleted values, indicating that the origins of river water were some spring waters recharging at higher elevations. This significant difference was probably caused by the amount of natural water in the river which was comparatively large, with respect to limited leachate discarded to the river. The value of δ^2H and $\delta^{18}O$ in shallow groundwater samples collected near to the river sites still represented local precipitation recharge, except at zone-3 monitoring well in Bantar Gebang landfill area indicated a mixing with leachate.

The difficulty to trace leachate movement to Cibitung River by using deuterium could be overcome by applying electrical conductivity parameter. The significant increasing conductivity values were detected in the river water closed to the landfills, it suggested that Cibitung River was influenced by leachate.

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