

**Tritium: Implies Young Groundwater Age?
Insight from the Isotope and Hydrochemical Data of Mud Volcano and Hydrocarbon
Well in East Java**

***Tritium: Mengimplikasikan Umur Airtanah Muda?
Pencerahan dari Data Isotop dan Hidrokimia Gunung Lumpur dan Sumur Hidrokarbon
di Jawa Timur***

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naskah diterima 10 Agustus 2021, selesai direvisi 01 November 2021, dan disetujui 22 November 2021

ABSTRACT

The use of tritium isotope is one of the methods for determining groundwater age. It can be used to determine the age of groundwater classified as young. However, if it is the only method used, the results may not be valid. In this study, tritium application in determining groundwater age was evaluated based on deuterium and oxygen-18 isotopes and hydrochemical data from seven mud volcanoes and one hydrocarbon production well in East Java Basin. The tritium analysis shows that the age of groundwater samples is young, 1.75 to 9 years. However, deuterium, oxygen-18, and hydrochemical analysis indicate that the groundwater age is relatively old. It shows that the results of groundwater age analysis using tritium are not valid in this study. It is because tritium is not only from the atmosphere but also from tritium enrichment below the surface through water and rock interaction. The shifting of oxygen-18 isotope, which becomes heavier, indicates that isotope enrichment occurred in the subsurface. Based on the composition of the major cations and anions, the groundwater samples in this study have Na-Cl type with high TDS values as saline water. The shifting of oxygen-18 isotope, the water type, and the high TDS value also indicate that water and rock interaction occurs beneath the surface and can increase the tritium content in groundwater.

Keywords: deuterium, groundwater age, hydrochemical data, mud volcano, oxygen-18, tritium

ABSTRAK

Salah satu metode untuk menentukan umur airtanah adalah dengan menggunakan isotop tritium. Tritium dapat digunakan untuk menentukan umur airtanah yang tergolong muda. Namun, jika hanya menggunakan metode ini, maka hasil yang diperoleh mungkin tidak valid. Dalam studi ini, penggunaan tritium untuk menentukan umur airtanah dievaluasi berdasarkan data isotop deuterium dan oksigen-18, serta data hidrokimia dari tujuh gunung lumpur dan satu sumur produksi hidrokarbon di Cekungan Jawa Timur. Analisis tritium menunjukkan umur sampel airtanah yang tergolong muda, yaitu 1,75 hingga 9 tahun. Namun, analisis deuterium, oksigen-18, dan analisis hidrokimia menunjukkan bahwa umur airtanah tergolong tua. Hal tersebut memperlihatkan bahwa hasil analisis umur airtanah berdasarkan tritium pada studi ini tidak valid. Hal ini disebabkan karena tritium tidak hanya berasal dari atmosfer, tetapi dapat juga berasal dari pengayaan tritium di bawah permukaan melalui reaksi air dan batuan. Pergeseran nilai oksigen-18 yang menjadi semakin berat mengindikasikan bahwa terjadi pengayaan isotop tersebut di bawah permukaan. Berdasarkan komposisi kation dan anion utama, sampel airtanah dalam studi ini memiliki tipe Na-Cl dengan nilai TDS yang tinggi dan air yang tergolong asin. Pergeseran oksigen-18, tipe air,

dan nilai TDS yang tinggi tersebut juga menunjukkan bahwa terjadi interaksi air dan batuan di bawah permukaan yang dapat meningkatkan nilai tritium pada airtanah.

Kata kunci: deuterium, umur airtanah, data hidrokimia, mud volcano, oksigen-18, tritium

INTRODUCTION

Groundwater age can be determined by using tritium isotope, which is a radioactive hydrogen isotope with a half-life of 12.32 years (Simpson, 1987). Tritium can be used for young groundwater age, *i.e.* less than 60 years, considering the increase of tritium in the atmosphere due to the nuclear test started in 1952 (Kazemi *et al.*, 2006). However, tritium application in determining groundwater age can cause uncertainty due to some reasons, such as mixing between young and old groundwaters and tritium enrichment below the surface (Kazemi *et al.*, 2006; Lindsey *et al.*, 2019). In this study, its use in determining groundwater age is evaluated based on deuterium and oxygen-18 isotopes and groundwater hydrochemical data from seven mud volcanoes and one hydrocarbon production well in East Java.

According to Milkov (2000), a mud volcano is a geological phenomenon that occurs almost worldwide, including in Indonesia. Sidoarjo mud volcano (LUSI), the largest mud volcano in the world (Mazzini, 2018), located in the East

Java Basin, is an example of mud volcano in Indonesia. Since its first initiation, the highest eruption rate of LUSI was 180,000 m³/day in December 2006 (Mazzini *et al.*, 2007). Beside LUSI, there are several other mud volcanoes in the East Java Basin, as shown in Figure 1. Based on Bouguer anomaly data, the mud volcanoes are generally located in the depression zone with a negative Bouguer anomaly, which indicates the deepest part of a basin.

Based on Davies *et al.* (2007) and Mazzini (2009), mud volcano generation has a strong relation with fluid pressure in rock layers. Due to geological processes, the fluid pressure increases and exceeds the rock strength, which causes fractures. As a result, fluid will flow through the fractures to the surface. The fluid will erode the rock layers it passes. Thus, it will come out to the surface as mud.

According to Li *et al.* (2014), the mud volcano fluid may be from some sources with a complex migration process. During the migration, the fluid chemical and isotope compositions may change

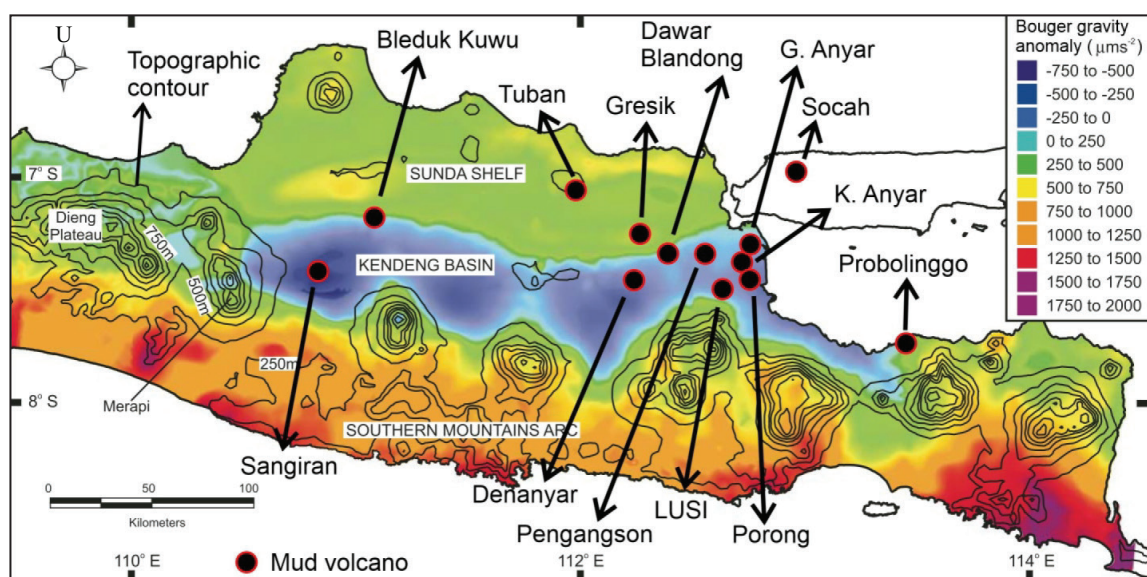


Figure 1. The Bouguer gravity anomaly map and the location of mud volcanoes in East Java (Modified from Smyth *et al.*, 2008 and Istadi *et al.*, 2009).

due to some processes such as water-rock interaction, organic material degradation, and fluid mixing between shallow and deep rock layers. Moreover, Lavrushin *et al.* (2005) suggested that evolution of isotopic composition can also occur as a part of a groundwater recharge system. Meanwhile, Mazzini *et al.* (2017) conducted geochemical and isotopic studies to compare mud volcanoes (including LUSI), cold springs, artesian wells, and volcano-hosted hydrothermal springs characteristics. The results show that LUSI is Na and Cl dominated, and its water is enriched in several mobile elements due to high-temperature fluid mineral interaction found in the subsurface.

The study area is located in the East Java Basin, which covers onshore and offshore areas of the eastern part of Java, as shown in Figure 2. Physiographically, the study area includes Rembang-Randublatung Zone, Dander High, and Kendeng Zone. Stratigraphically (Figure 3), overpressured rock layers are found in the formations with thick shale layers, such as Tuban Formation and Kujung Formation (Ramdhan *et al.*, 2013). The structures which generally

develop in the East Java Basin consist of west-east faulted anticline, as can be seen in the cross section of Figure 2.

METHODS

The methods used in this study consist of groundwater isotope and hydrochemical analysis. Isotope analysis conducted consists of tritium, deuterium, and oxygen-18, while hydrochemical analysis conducted includes total dissolved solids (TDS), anion and cation (carbonate alkalinity, chloride, sulfate, calcium, magnesium, potassium, and sodium). Groundwater sampling for the analysis was carried out at the eight points in September 2015, as shown in Figure 4. All water samples were filtered using 0.2 μm filters. Before sampling, bottles used (polyethylene and glass bottles) were rinsed with water from each sampling location. Water samples for cation analysis were preserved by acidifying with nitric acid until the pH was less than 2. Meanwhile, water samples for anion and isotope analysis were unpreserved. The data availability is shown in Table 1. Mean-

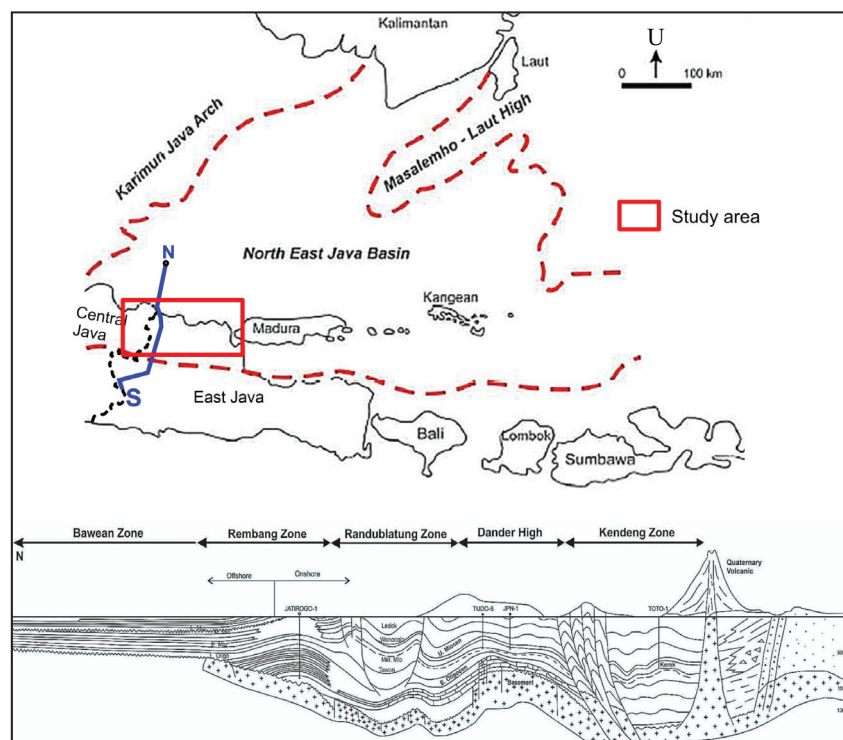


Figure 2. The boundary and physiography of East Java Basin (Modified from Pertamina BPPKA, 1996 and Ramdhan *et al.*, 2013).

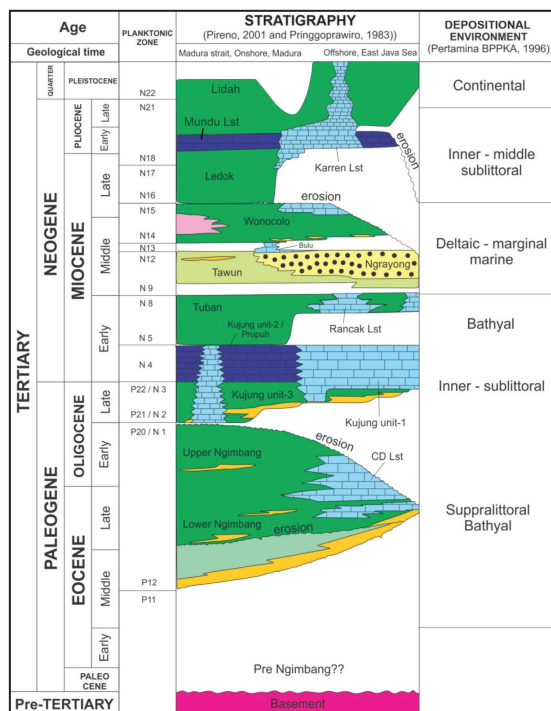


Figure 3. Stratigraphic column of East Java Basin (modified from Pringgoprawito, 1983; Pertamina BPPKA, 1996; Mujiono and Pireno, 2001).

while, Figure 5 shows a documentation example of the sampling.

The analysis of tritium, deuterium, and oxygen-18 isotopes were used to estimate and evaluate the age and source of groundwater. Groundwater age calculation using tritium was conducted

based on rainwater tritium data in Kampar, Riau (Musti'atin, 2015). The tritium composition of the rainwater was 3.95 TU (Tritium Unit). Tritium content on groundwater samples is calculated using the following equation (Clark, 2015):

$$t = -17.77 \ln(T_t/T_0) \dots\dots\dots (1)$$

where:

t : Groundwater sample age

T_0 : Initial tritium content (tritium in rainwater)

T_t : Tritium content in the groundwater sample

Beside evaluating groundwater age, deuterium and oxygen-18 isotopes were also used to interpret the mechanism that controlled the isotope composition of the groundwater samples. Meanwhile, the hydrochemical analysis was carried out to determine the water type of groundwater samples based on the plot of anion and cation on the Piper diagram (Piper, 1944).

RESULTS AND DISCUSSION

The results of tritium isotope analysis of seven groundwater samples from mud volcanoes and the hydrocarbon production well can be seen in Table 2. It shows that the tritium in groundwa-

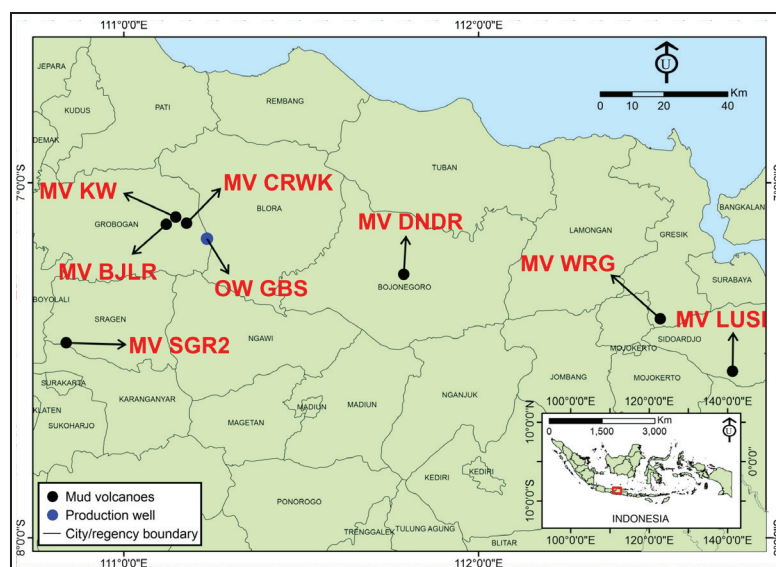


Figure 4. Map of groundwater sampling location from mud volcanoes and hydrocarbon production well.

Table 1. The data availability in this study

No	Samples	Types	Locations	Tritium Data	Deuterium and Oxygen-18 Data	Hydrochemical Data
1	MV WRG	mud volcano	Gresik	✓	✓	✓
2	MV CRWK	mud volcano	Grobongan	✓	✓	×
3	MV KW	mud volcano	Grobongan	✓	✓	×
4	OW GBS	production well	Grobongan	✓	✓	✓
5	MW LUSI	mud volcano	Sidoarjo	✓	✓	✓
6	MV DNDR	mud volcano	Bojonegoro	✓	✓	×
7	MV SGR2	mud volcano	Sragen	✓	✓	✓
8	MV BJLR	mud volcano	Grobongan	×	×	✓
✓ : available						
×						
: not available						



Figure 5. A documentation example of groundwater sampling of mud volcano in Banjarlor (code: MV BJLR in Figure 4).

ter samples ranges from 2.38 to 3.58 TU, with groundwater age which ranges from 1.75 to 9 years, classified as young groundwater based on Kazemi *et al.* (2006). Young groundwater is commonly found as meteoric water, shallow groundwater, or surface water, but not common for deep groundwater as analyzed in this study. The result of deuterium and oxygen-18 isotope analysis is shown in Figure 6. It shows the plot

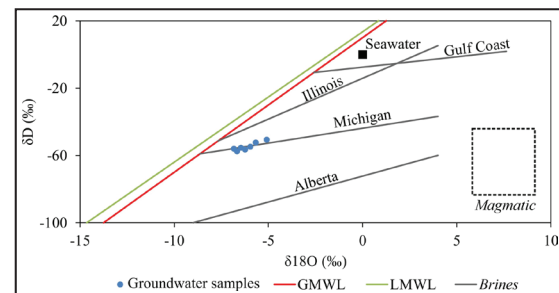


Figure 6. The plot of deuterium and oxygen-18 isotopes of groundwater samples against local meteoric water line (LMWL), global meteoric water line (GMWL), seawater (Clark, 2015), magmatic water (White, 1974 as cited in Nicholson, 1993), and brines (Clayton *et al.*, 1966).

of groundwater isotope of the mud volcanoes and production well compared with the local meteoric water line in Sidoarjo area (Satrio *et al.*, 2012) and the global meteoric water line (Craig, 1961). Based on the plot, the groundwater samples do not show the magmatic water or seawater characteristics, but having isotope compositions that fall on linear regression line of formation water isotopes from the Michigan Basin studied by Clayton *et al.* (1966). The plot also shows that there is oxygen-18 shifting around +2.1

Table 2. The results of tritium, deuterium, and oxygen-18 isotopes analysis

No	Samples	Tritium Unit (TU)	Groundwater Age (years)	Deuterium (‰)	Oxygen-18 (‰)
1	MV WRG	3.58	1.75	-56	-6.84
2	MV CRWK	3.24	3.52	-55.4	-6.47
3	MV KW	2.71	6.70	-52.2	-5.67
4	OW GBS	3.52	2.05	-57.5	-6.67
5	MW LUSI	3.05	4.59	-54.7	-5.96
6	MV DNDR	2.38	9.00	-50.5	-5.09
7	MV SGR2	3.4	2.66	-56.5	-6.25

to +3.1 from the local meteoric water line. It indicates that there was oxygen-18 isotope enrichment due to water-rock interaction. If there is no enrichment, the plot of oxygen-18 isotope should have fallen around the local meteoric water line. Thus, it implies that the age of the groundwater samples is older than the recent meteoric water.

The result of hydrochemical analysis of groundwater samples from four mud volcanoes and the production well is shown in the Piper diagram in Table 3 and Figure 7. All the groundwater samples analyzed have Na-Cl type. Table 3 shows the TDS values of groundwater samples, ranging from 13,235 to 47,472 mg/L.

The maximum TDS value is higher than the approximate seawater TDS, *i.e.*, 35,000 mg/L (Freeze and Cherry, 1979). According to Freeze and Cherry (1979), the TDS values show that the groundwater is saline. The high TDS values with dominant ion composition of Na and Cl are the characteristics of formation water and indicate that water-rock interaction has occurred for a long time (Chebotarev, 1955). This result also supports our hypothesis about the enrichment of the oxygen-18 isotope.

Compared with the deuterium and oxygen-18 isotopes and the hydrochemical analysis results, the groundwater age from tritium isotope analysis may not represent the actual

Table 3. The results of anion and cation analysis of groundwater samples

No.	Parameters	Units	MV BJLR	MV SGR2	MV WRG	MV LUSI	OW GBS
1	TDS	mg/L	47472	16648	27432	42476	13235
	Anions						
2	Carbonate Alkalinity as CaCO ₃	mg/L	<1	<1	<1	<1	<1
3	Chloride, Cl ⁻	mg/L	28300	10600	18000	26900	8490
4	Sulfate, SO ₄ ²⁻	mg/L	77	<2	28	263	<2
	Cations						
5	Calcium, Ca	mg/L	511	153	<0.05	1270	18.1
6	Magnesium, Mg	mg/L	93.8	117	117	416	107
7	Potassium, K	mg/L	389	56.0	86.3	227	56.9
8	Sodium, Na	mg/L	18100	5720	9200	13400	4560

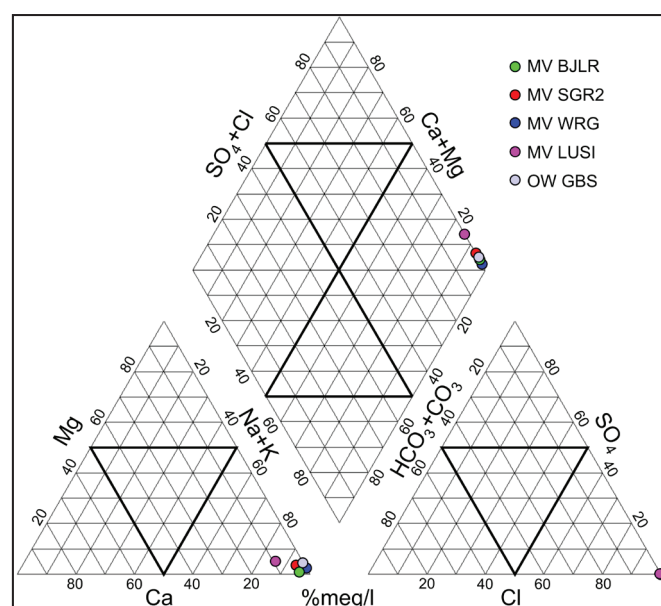


Figure 7. The plot of anion and cation composition of groundwater samples on the Piper diagram.

groundwater age. As a comparison, Satrio *et al.* (2012) also showed that the groundwater age from LUSI analyzed a year after the tragedy was old, older than 40,000 years. The young groundwater age from tritium analysis may be due to tritium enrichment in the atmosphere or below the surface (Kazemi *et al.*, 2006). However, since the groundwater samples in this study represent the deep groundwater from the overpressured zone (Ramdhan *et al.*, 2013), it is less likely that the tritium enrichment occurs in the atmosphere.

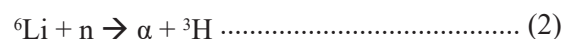
According to Lehmann *et al.* (1993), tritium isotope in deep groundwater may be caused by four factors such as contamination of groundwater by drilling fluid which contains high tritium, the presence of a highly permeable zone that accommodates tritium movement from the surface to the deep groundwater zone, measurement error, and tritium generation below the surface.

In this study, the first factor can not be analyzed because there is no data concerning tritium composition in drilling fluids used during the drilling in the study area. Based on Ramdhan *et al.* (2013), the fluid pressure of the shallow groundwater zone in the study area is hydrostatic. Meanwhile, in deeper depth, the fluid is in an overpressure condition. It indicates that there is a confining layer that separates the hydrostatic and overpressure zones. Therefore, it is less likely that there is a permeable zone that connects the atmosphere with the deep groundwater. Thus, the second factor of tritium occurrence in deep groundwater can also be ruled out. The possibility of measurement error can also be eliminated because the measurement results show relatively consistent tritium values. Therefore, the possible factor responsible for the tritium enrichment is tritium generation below the surface.

Many reactions can generate tritium below the surface (Davis and Murphy, 1987 as cited in Lehmann *et al.*, 1993). However, only the lithium reaction can generate tritium signi-

ficantly (Andrews and Kay, 1982). Lithium has two stable isotopes, *i.e.* lithium-6 and lithium-7, each with abundance of 7.5 % and 92.5 %, respectively (Liu *et al.*, 2018). According to Tang *et al.* (2007), lithium isotope fractionation occurs significantly in the low-temperature system but will become various in altered oceanic crust, hydrated mantle rocks, and eclogitic slabs.

Lithium is relatively conservative despite its high solubility because it is not involved either in redox or biological reactions. The reactions of lithium-6 and lithium-7 isotopes resulting in tritium occur as the two following equations:



Therefore, the relatively high tritium content in the groundwater samples in the study area may be caused by the tritium enrichment from lithium-rich rocks.

Tang *et al.* (2007) created a diagram that shows lithium-7 isotope composition in nature as shown in Figure 8. Based on the stratigraphy of the study area (Figure 3), the subsurface lithology is dominated by limestone and shale deposited in marine environment, with blue and green colors in the stratigraphic column, respectively. By referring to Figure 8, there are two possible sources of lithium-7 that can increase the tritium isotope composition in deep groundwater due to water-rock interaction, *i.e.* carbonate deposits with lithium-7 composition ranging from +14 to +40 ‰ and marine deposits with lithium-7 composition ranging from -1 to +25 ‰. Moreover, Mazzini *et al.* (2017) suggested that interaction between hydrothermal fluids and rock facilitated lithium enrichment in the fluids. This process is expected to occur not only for a long time but also continuously maintain the tritium isotope composition in groundwater samples of this study.

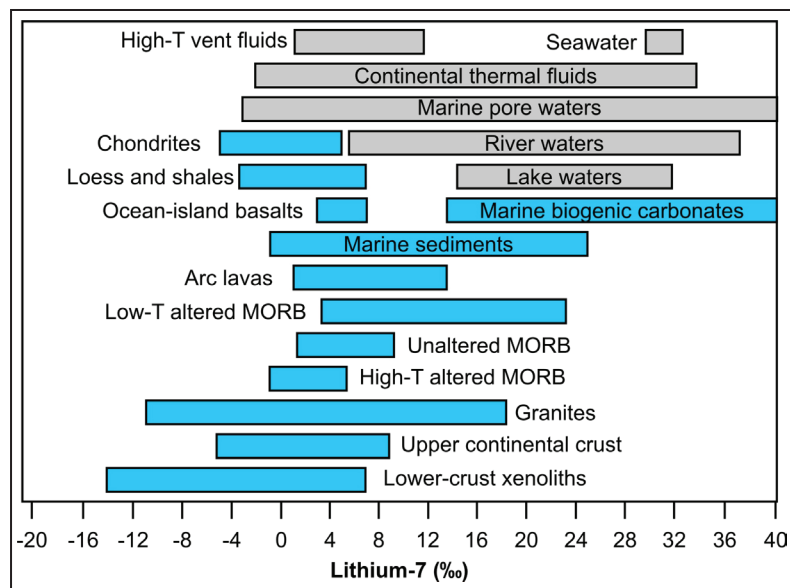


Figure 8. The composition of lithium-7 isotope in nature (modified from Tang *et al.*, 2007).

CONCLUSIONS

The results of deuterium and oxygen-18 isotopes and hydrochemical analysis indicate that the age of groundwater samples from the mud volcanoes and hydrocarbon production well in this study is old (more than 60 years) due to intensive water-rock interactions, even though the tritium isotope analysis shows the opposite, that the groundwater age is young. It indicates that the groundwater age from the tritium analysis is not valid in this study because the tritium isotope source is not only from the atmosphere but also from tritium enrichment below the surface through the interaction of water and rocks with high lithium-6 and lithium-7 content. Therefore, the use of tritium isotope in determining groundwater age should be conducted carefully and validated using other methods, such as deuterium and oxygen-18 isotopes, hydrochemical analysis, and another multi tracer dating.

ACKNOWLEDGEMENT

The authors thank SKK Migas for organizing the study of overpressure in Indonesia and PT. Pertamina EP for the funding of this study.

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