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Identifying the sources of produced water in the oil field by isotopic techniques

Nguyen Minh Quy, Hoang Long, Le Thi Thu Huong, Luong Van Huan
Vietnam Petroleum Institute, 173 Trung Kinh, Yen Hoa,Cau Giay, Ha Noi
Vo Thi Tuong Hanh
Hanoi Irradiation Center, Vietnam Atomic Energy Institute
Km12, Minh Khai, Nam Tu Liem, Ha Noi

Abstract: The objective of this study is to identify the sources of the formation water in the Southwest Su Tu Den (STD SW) basement reservoir. To achieve the objective, isotopic techniques along with geochemical analysis for chloride, bromide, strontium dissolved in the water were applied. The isotopic techniques used in this study were the determination of water stable isotopes signatures (δ^2H and δ^{18}O) and of the ^{87}Sr/^{86}Sr ratio of strontium in rock cutting sample and that dissolved in the formation water. The obtained results showed that the stable isotopes compositions of water in the Lower Miocene was -3‰ and -23‰ for δ^{18}O and δ^2H, respectively indicating the primeval nature of seawater in the reservoir. Meanwhile, the isotopic composition of water in the basement was clustered in a range of alternated freshwater with δ^{18}O and δ^2H being –(3-4)‰ and -(54-60)‰, respectively). The strontium isotopes ratio for water in the Lower Miocene reservoir was lower compared to that for water in the basement confirming the different natures of the water in the two reservoirs. The obtained results are assured for the techniques applicability, and it is recommended that studies on identification of the flow-path of the formation water in the STD SW basement reservoir should be continued.

Keywords: water stable isotopes composition, strontium isotopes ratio, the sources of produced water

I. INTRODUCTION

Deuterium (^{2}H) and oxygen-18 (^{18}O) are environmental isotopes and they are the constituents of natural water molecules. The deuterium and oxygen-18 in the notation of δ^2H and δ^{18}O are widely used as the signature in studies of the geochemical evolution, recharge processes and water-rock interaction [1, 2, 3].

Strontium is a minor component in the formation water. It is a reactive soluble cation that readily substitutes for Ca^{2+} in carbonates, sulfates, feldspars and other rock-forming minerals. Strontium participates in water-rock interactions. The element is the ninth most abundant constituent dissolved in seawater, and it behaves geochemically like calcium.

In basement rocks the abundance of ^{87}Sr, a daughter of ^{87}Rb, is directly linked to the geochemistry of potassium. Therefore, the ^{87}Sr/^{86}Sr ratio is usually characterized for the geochemistry and age of rocks. With Oligocene and Miocene sedimentary rocks the ^{87}Sr and ^{86}Sr content in grain matrix is inherited from parent rocks, while the content of ^{87}Sr and ^{86}Sr in the joints and cements (mainly secondary carbonate) is possibly from the equilibrium with those of formation water.

Formation water discharged into the

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aquifer for a certain long time (in sense of geologic time), so that water-rock interaction could be taken place for a period enough to achieve isotopic equilibrium among minerals in the formation water and aquifer host rocks. In this case the $^{87}Sr/^{86}Sr$ ratio is the most important indicator for the interaction equilibrium. Clastic sediment rocks usually have well-recognized pattern of the $^{87}Sr/^{86}Sr$ isotope ratio in leachates or calcareous minerals according to Strontium Isotope Stratigraphy (SIS) [4, 5], while igneous rocks contain minerals which usually have distinct Sr content and isotope ratio related to mineral type and age of rocks. The SIS curve characterizing for the geologic age of minerals was constructed based on the $^{87}Sr/^{86}Sr$ ratios in typical marine carbonates. This may be done because the oceanic recycling rate is relatively short (<1000 years) compared to the long oceanic residence time of strontium (2–4 Ma) and no natural fractionation of stable strontium isotope was observed during natural processes. This property makes the strontium isotopic ratio to be a reliable candidate for tracing strontium origin, for evaluating mixing of ground waters and for studying a state of isotope of equilibrium between groundwater and strontium bearing minerals and rocks.

Understanding primeval nature of water before discharging into aquifer by isotopic signatures, in combination with tracing water-rock interaction by comparing isotope equilibrium, can help finding the sources of production water in a well field.

The formation waters from the oil production well field Black Lion southwest (STD SW), due to the low strontium concentration in the potential primeval waters, water-rock interaction may approach isotopic equilibrium with respect to strontium. This process could be identified by comparing the $^{87}Sr/^{86}Sr$ values of the primary minerals of the host rock and that of the secondary minerals on the joints, cements and pores with those of the strontium dissolved in the formation water.

The purpose of this study was to identify the sources of produced water in the oil well fields of the Black Lion southwest by using isotopic signatures of produced water δ^2H and $\delta^{18}O$) and the $^{87}Sr/^{86}Sr$ ratio of minerals from the fields.

II. EXPERIMENTAL

The isotopic compositions of the produced water were measured on an isotopes ratio mass spectrometer, Micro Mass (UK) equipped with an elemental analyzer (Euro Vector, Italy) to convert water molecules into hydrogen and carbon dioxide suited for the deuterium and oxygen analysis. The water stable isotope compositions are defined the differences of the deuterium to protium and oxygen-18 to oxygen-16 ratios in a sample from those of the Vienna Standard Mean Ocean Water (VSMOW). These differences were expressed by the notation δ^2H and $\delta^{18}O$ in per mill (‰) as follows:

$$\delta^2 H = \left(\frac{^2R_{sample}}{^2R_{std}} - 1\right).1000$$

$$\delta^{18} O = \left(\frac{^{18}R_{sample}}{^{18}R_{std}} - 1\right).1000$$

Where $^2R_{sample}$, $^2R_{std}$, $^{18}R_{sample}$ and $^{18}R_{std}$ are the ratio of deuterium to protium and oxygen-18 to oxygen-16 in sample and in the VSMOW standard, respectively.

The pattern, in which $\delta^{18}O$ and δ^2H deviated from the Global Meteoric Water Line (GMWL) that was described by an aquation as:

$$\delta^2H = 8\delta^{18}O + 10, \text{‰ } vs.\text{VSMOW [3]}$$

was used to trace the geochemical evolution or water-rock interaction that formation water has been involved, and then interpret the primeval nature of the formation water.

The $^{87}Sr/^{86}Sr$ isotope ratios in formation water and rock cutting samples are measured using the Thermo Scientific™ TRITON™ Plus Multicollector thermal ionization mass spectrometric (TIMS,) method and given directly as atomic mass ratios. The $^{87}Sr/^{86}Sr$ ratio was analyzed for bulk samples of the STD basement rock and for both leachates and residues of Oligocene and Miocene sedimentary rocks. The leachates were prepared by dissolution of the sediment with acetic acid according to a standard procedure described elsewhere [**7, 8**]. Briefly, it was as follows: allow 0.3g sample to leach in 5 mL solution of CH_3COOH 4M in a glass tube. The content in the tube was intermittently shaken for 8h at the room temperature, then allowed to settle for 16 hours so that the total leaching time was 24h. The leachate was separated from the residues by centrifugation. The strontium isotopes ratio in the residues and leachates were determined by the TIMS method as described above.

With this extraction procedure, the leachates are considered to be a solution of the secondary carbonate which mainly constitutes the joints and cements, while the residues being the grain matrix of sediments.

III. RESULTS AND DISCUSSIONS

Analytical results

Tables I presents the results of the isotopic composition of produced water from the lower Miocene and basement reservoirs. Table II shows the results of the $^{87}Sr/^{86}Sr$ ratio in strontium dissolved in produced water of the two above mentioned formations. In Table II the choride concentration in the salinity unit (part per thousand, ppt) in the produced water is also shown. Table III presents the ratios of $^{87}Sr/^{86}Sr$ in the acetic acid leachates and residues of the Miocene and Oligocene rock cutting minerals.

Table 1. The ^{18}O and ^{2}H isotopic composition of water samples taken from the lower Miocene and basement reservoirs

No.	Reservoir	Well	Acquisition Date	$\delta^{18}O_{VSMOW}$ (‰)	$\delta^{2}H_{VSMOW}$ (‰)
1	Lower Miocene	SD-11P	18-Jul-10	-3.03	-24.48
2	Lower Miocene	SD-20P	5-Jul-10	-3.18	-23.30
3	Lower Miocene	SD-27P	18-Jul-10	-2.85	-23.07
4	Basement	SD-3P	18-Jul-10	-3.43	-57.52
5	Basement	SD-4PST	18-Jul-10	-4.09	-53.61
6	Basement	SD-5PST	3-Jul-10	-4.52	-59.76
7	Basement	SD-6P	3-Jul-10	-3.98	-56.30
8	Basement	SD-7P	18-Jul-10	-3.79	-57.37
9	Basement	SD-18P	3-Jul-10	-3.48	-58.70
10	Basement	SD-19P	3-Jul-10	-3.61	-60.07
11	Basement	SD-22P	18-Jul-10	-3.66	-59.92

Table II. The $^{87}Sr/^{86}Sr$ ratio of strontium dissolved in water in the lower Miocene and basement reservoirs

No.	Reservoir	Well	Acquisition Date	$^{87}Sr/^{86}Sr$	Sr (mg/L)	Chloride (ppt)
1	Lower Miocene	SD-11P	18-Jul-10	0.707571	3.39	30.09
2	Lower Miocene	SD-20P	5-Jul-10	0.707554	3.27	29.65
3	Lower Miocene	SD-27P	18-Jul-10	0.707610	3.02	29.87
4	Basement	SD-3P	18-Jul-10	0.706884	9.65	10.19
5	Basement	SD-4PST	18-Jul-10	0.706845	10.24	12.42
6	Basement	SD-5PST	3-Jul-10	0.706900	8.21	9.43
7	Basement	SD-6P	3-Jul-10	0.706833	7.87	19.19
8	Basement	SD-7P	18-Jul-10	0.706901	8.67	13.03
9	Basement	SD-18P	3-Jul-10	0.706865	7.84	11.48
10	Basement	SD-19P	3-Jul-10	0.706839	8.63	10.58
11	Basement	SD-22P	18-Jul-10	0.706901	7.38	10.47

Table III. The $^{87}Sr/^{86}Sr$ isotope ratio of strontium in the acetic acid leachates and minerals of the Miocene and Oligocene rock cutting samples

Reservoir	**Well**	**Depth (mTVD)**	**$^{87}Sr/^{86}Sr$ (Residue)**	**$^{87}Sr/^{86}Sr$ (Leachate)**	**Remark**
Miocene	SD-26P	1722	0,7112537	0,7076215	B10/ BH Formation
Miocene	SD-26P	1723	0,7113246	0,7075875	B10/ BH Formation
Miocene	SD-26P	1725	0,7111897	0,7075817	B10/ BH Formation
Miocene	SD-27P	1720	0,7108979	0,7107257	Oil Zone/ Miocene Tuff
Miocene	SD-27P	1721	0,7112812	0,7105179	Oil Zone/ Miocene Tuff
Oligocene	SD-4P	2228	0,7108124	0,7088826	C30
Oligocene	SD-4P	2237	0,7105152	0,7088822	C30
Oligocene	SD-4P	2246	0,7114355	0,7091526	C30
Oligocene	SD-1X	2162	0,7107647	0,7085093	C30
Oligocene	SD-1X	2171	0,7105561	0,708493	C30

Isotope and geochemical characterizations of water in the Miocene and basement formation are given in Table IV and V, respectively.

Table IV. Characterizations of water in the Miocene formation taken for this study

Well	Acquisition Date	Chloride (ppt)	Bromide (mg/L)	$\delta^{18}O_{VSMOW}$ (‰)	$\delta^{2}H_{VSMOW}$ (‰)
SD-10P	25-Aug-10	30.17	79.2		
SD-10P	13-Oct-10	28.06	70.5		
SD-11P	18-Jul-10	30.09		-3.03	-24.48
SD-20P	5-Jul-10	29.65		-3.18	-23.30
SD-27P	18-Jul-10	29.87		-2.85	-23.07

Table V. Chloride and isotopic composition of Basement water

Well	Acquisition Date	Chloride (ppt)	$\delta^{18}O_{vsmow}$ (‰)	$\delta^{2}H_{vsmow}$ (‰)
SD-3P	18-Jul-10	10.19	-3.43	-57.52
SD-4PST	18-Jul-10	12.42	-4.09	-53.61
SD-5PST	3-Jul-10	9.43	-4.52	-59.76
SD-6P	3-Jul-10	19.19	-3.98	-56.30
SD-7P	18-Jul-10	13.03	-3.79	-57.37
SD-18P	3-Jul-10	11.48	-3.48	-58.70
SD-19P	3-Jul-10	10.58	-3.61	-60.07
SD-22P	18-Jul-10	10.47	-3.66	-59.92

Primeval Sources of the Formation Waters

The origin of the formation waters in high-temperature deep-geological structure can be distinguished by their geochemical and isotopic compositions. Commonly, water in geological deep structures can be originally recharged from the following major water sources [2, 3].

+ Andesitic water: this source is recycled seawater which has been sub-ducted in regions with andesitic volcanism or convergent plate volcanism. The $\delta^{18}O$ and $\delta^{2}H$ values of this water are about +(10 ± 2)‰ and +(20 ± 10)‰, respectively, while the chloride concentration is similar to that of the seawater;

+ Juvenile water: this source is water coming from the Earth's mantle or core that has never been a part of the hydrospheric cycle;

+ Magmatic water: this source is water that was in equilibrium with magma, regardless of its initial origin. The only separated aqueous phase is magmatic water but not water physically dissolved in the magma;

+ Meteoric water: This water is originated from precipitation and of any age (rain, snow, ice, river water, lake water, groundwater) (Fig.1);

+ Ocean water or seawater: this is water in open oceans which may enter the deep-geological structure.

Andesitic water, juvenile water and magmatic water (sometimes called "tectonic water") are greatly different from seawater and meteoric water. They are easily recognized by the geochemical compositions, e.g. SiO_2, Ca^{2+}, Mg^{2+}, Ba^{2+}, Li^+, K^+, CO_3^{2-}, S^{2-}, Cl^-, F^-, etc. as well as the isotopic composition, i.e. δ^{18}O and δ^2H. The former group of water usually have high content of solute silica, gases. They are also much enriched with heavy isotopes of water, i.e. the values of δ^{18}O are positive (> +2‰) due to the extensive ^{18}O isotope exchange or equilibration with parent rocks.

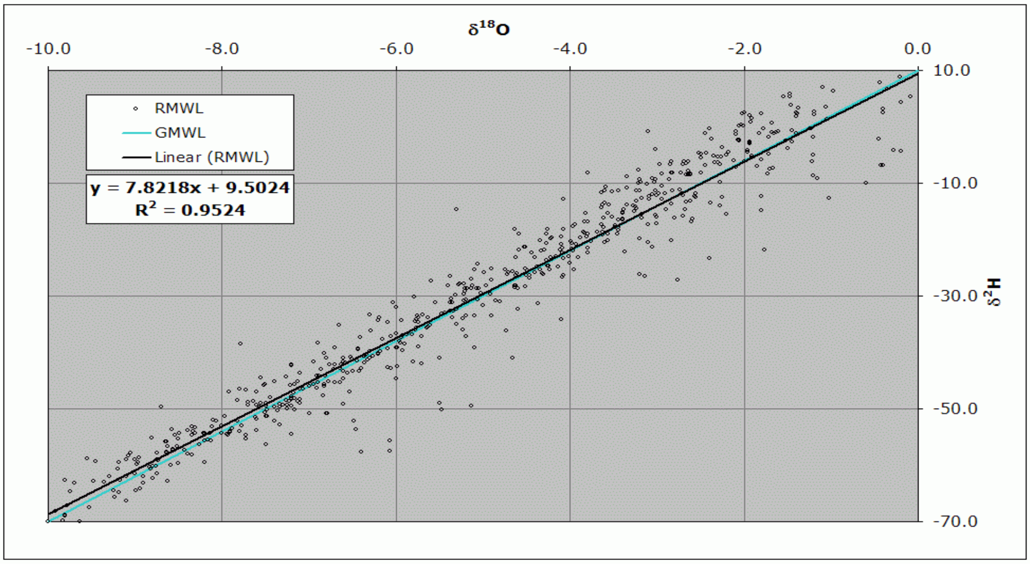

Fig.1: The GMWL ($\delta^2H = 8\delta^{18}O + 10$) is plotted with the regional meteoric water line (RMWL: $\delta^2H = 7.82\delta^{18}O + 9.50$, black colour line) constructed from observed data around the South East Asian region (dots)

In this study the waters in the Miocene and basement reservoirs from the southwest Black Lion (STD SW) oilfield are of concerns. From Fig 1 and Table 1 it was revealed that there is no evidence for the occurrence of tectonic water in the Miocene and Basement reservoirs. The primeval nature of the STD formation waters might be regarded to as seawater and/or meteoric water because all the □^{18}O values of water in the reservoirs are negative (Table 1). Meteoric water has its δ^{18}O and δ^2H evolutes according to Meteoric Water Line (MWL) and this line has the clear global (GMWL) or regional (RMWL) pattern (Fig 1) [3].

Figure 2 depicts the typical positions of concurrent fresh groundwater, seawater and paleowater in relation to Global and Regional Meteoric Water Lines. In aquifers with fresh groundwater which are locally recharged with recent precipitation, the compositions of the heavy isotopes are commonly similar to the annual mean of these quantities in the rainfall for that area. Therefore, the δ^{18}O and δ^2H of recent groundwater are aligning close to the RMWL. The δ^{18}O and δ^2H of seawater should be clustered around the zero point of the ordinate (by the definition of the notation). Seawater signature is also easily recognized by comparing the Cl^-, Br^- concentration with the δ^{18}O value.

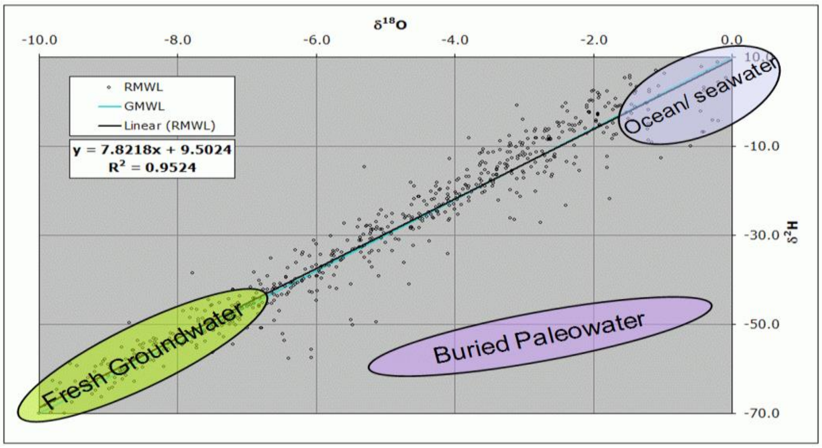

Fig.2. Typical area of fresh groundwater, seawater and paleowater in relation to the Global and Regional Meteoric Water Line

Paleowater is buried and experienced with long and extensive water-rock interactions or high-temperature steam loss, therefore, its $\delta^{18}O$ and $\delta^{2}H$ is largely shifted from the GMWL or RMWL (Fig 3 and Fig 4).

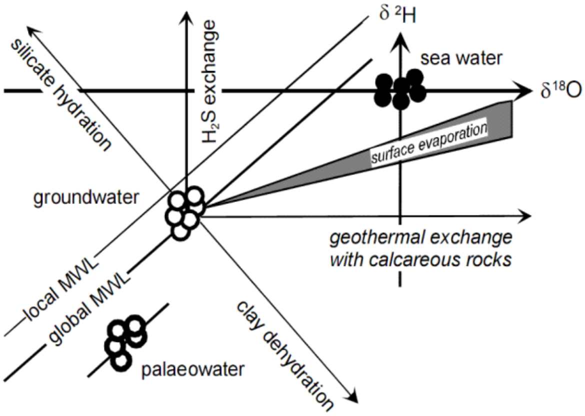

Fig.3. Various water-rock interaction processes which shift the $\delta^{18}O$ and $\delta^{2}H$ values from the MWL (adapted from [3])

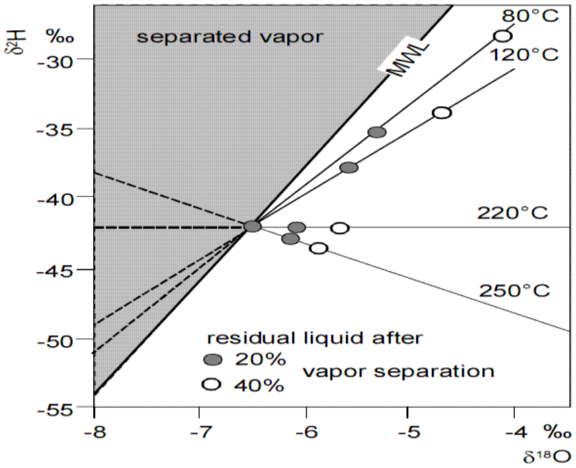

Fig.4. The $\delta^{18}O$ and $\delta^{2}H$ shifted from the MWL due to the steam loss at various high temperatures (adapted from [3])

It can be seen from Table 4, that the chloride of water in the Miocene formation is highly homogenous despite the water were sampled from different wells and at greatly different times. These water samples also have a close correlation of chloride and bromide concentration which is higher than that of today seawater (the results not shown here). In this study, the δ^{18}O and δ^{2}H values of water in the Miocene formation are positioned below the RMWL but somewhat close to the area of seawater (Fig 5). However, the values of □^{2}H and □^{18}O are more negative than that in seawater (red dots cluster in Fig 5). With all the mentioned above evidences, it might be suggested that the primeval nature of water in the Miocene formation is ancient seawater.

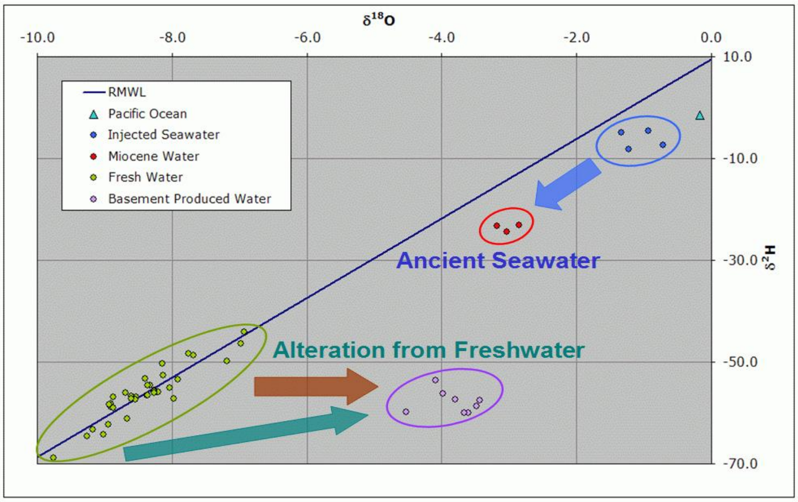

Fig.5. The δ^{18}O and δ^{2}H values of the waters from the STD SW Miocene & basement reservoir and their predicting primeval nature

The δ^{18}O and δ^{2}H in produced water from the basement shown in Table 1 and plotted in Fig 5 also indicates a clear pattern of paleo-groundwater (open dots cluster in Fig5). The δ^{2}H values in the produced water from the basement characterize for fresh water (from -57 to -60‰), while the δ^{18}O values are in the range of seawater (around -4‰). However, the chloride of water in the basement is far lower than that of the seawater. It was of 10-12 ppt *versus* 30 ppt in seawater (Table 1). These signatures clearly indicate that the primeval nature of water in the basement formation is meteoric but the water has undergone the isotope alterations by thermal water-rock interactions (the brown arrow in Fig 5, see also Fig 3) and/or steam separation processes under high temperature (green arrow in Fig 5, see also Fig 4). The conclusion on the primeval meteoric origin of water in the basement formation agrees with the results of our previous study on STD SW basement produced water.

Strontium Isotope Equilibrium

It can be seen from Table 2 that despite water samples were taken from various wells, but the ^{87}Sr/^{86}Sr isotope ratios of strontium dissolved in water of the basement formation are all close to each other being around 0.7068-0.7069. Three Miocene formation water samples have close ^{87}Sr/^{86}Sr isotope ratios of around 0.7075-0.7076 (Table 2). It should be noted that the instrumental precision in the ^{87}Sr/^{86}Sr analysis must be reached 6 decimal numbers as difference at the 4th decimal number could be a great value accounting for an uncertainty of millions years in the geologic scale. So, the $^{87}Sr/^{86}Sr$ ratios found for strontium dissolved in water in the Miocene and basement reservoir has distinct $^{87}Sr/^{86}Sr$ values indicating the different nature of water in the corresponding formations. The $^{87}Sr/^{86}Sr$ ratios of strontium in the residues of Miocene and Oligocene clastic sediment rocks are in a range of 0.7105 to 0.7113 and higher than that in seawater (Table 3). The strontium isotopes ratios found in the residues are largely distinctive from the corresponding quantity in the leachates, ranging from 0.7083 to 0.7107 (Table 3). These differences might be found in the nature of strontium in the two fractions. On the one hand, the residues are inherited from parent rocks having older age and as the consequence the ^{87}Sr contribution from the long-live ^{87}Rb decays should be higher. On the other hand, the residue components, virtually being solid grain matrix, are isolated or have limit possibility to contact with pore fluids, so they might have no interaction with the formation water leading to negligible effects on the $^{87}Sr/^{86}Sr$ ratios in the formation water. Moreover, sediment grains remained after long processes of weathering, erosion and transportation from distant parent rocks, are usually chemically stable and inert towards the interaction with intact water. All these made the $^{87}Sr/^{86}Sr$ ratios in the residues to be higher compared to those in leachates.

The data of the $^{87}Sr/^{86}Sr$ ratios clearly shows the equilibrium of strontium isotopes between Miocene formation water and the leachates from siltstone of the Miocene B10 sequence. In the previous section, we have concluded on the primeval nature of the Miocene formation water. This primeval water source has its typical strontium behavior and the predictable fashion of $^{87}Sr/^{86}Sr$ ratio. In our previous studies on sedimentary facies it has been indicated, that Miocene clastic sediments were formed in marine environments and the formation water, which was discharged in these sediments, is likely at an age of Miocene or later geologic time. Accordingly, the original $^{87}Sr/^{86}Sr$ ratio of Miocene formation seawater can be predicted by lookup from the Strontium Isotope Stratigraphy (SIS) curve [4, 5] and the predicted value should be greater than 0.7082. The $^{87}Sr/^{86}Sr$ value found in this study was approximately 0.7076 for the Miocene formation water. Based on the data found and the SIS curve, the formation water in the Miocene would be dated back to the Late Cenozoic to Paleogene age of about 75 Ma. It is unlikely to happen because this was an erosion period in the study area. Therefore, the $^{87}Sr/^{86}Sr$ ratios of 0.7075-0.7076 in the Miocene formation water are possibly indicated on the controlling of the equilibration with the solid phase in the joints and cements of Miocene clastic sediments which have similar $^{87}Sr/^{86}Sr$ ratios of approximately 0.7076 in the leachates.

There are also two samples taken in the top of the Miocene B10 layer of well 27P and described as volcanic tuffs in the petrology analysis report [6] of which their leachates have quite different $^{87}Sr/^{86}Sr$ ratios in comparison with those in the leachates from siltstone of Miocene B10 (Table 3). Volcanic tuffs, if really existed at the Miocene B10 top, might be local lens and hardly interact with formation water and contributed to the strontium isotope equilibrium. It is also likely that these samples are unrepresentative for Miocene clastic sediment as perceived by the description in the well 27P petrology report [6].

The $^{87}Sr/^{86}Sr$ ratios in basement produced water being around 0.7068-0.7069, are relatively low and distinct from those in the Miocene and Oligocene cutting rock. These results have excluded Oligocene sediments as the potential host aquifers for basement formation water. This because of that the primeval source of STD SW basement formation water was the meteoric. This kind of water originally contains very low concentration of strontium, eg. Sr concentration in river water typically is around 0.6 mg/L, and that in circular groundwater is around 1 mg/L. However, the Sr concentration found in all the basement water samples was higher than seawater of 8 mg/L. These results clearly indicate that strontium concentration in basement water was contributed from the dissolution in host rocks of aquifers or along flow-path. These dissolution processes assure the $^{87}Sr/^{86}Sr$ ratio in formation water to be an indicator for those ratio of aquifer host rocks. A broader collection of the $^{87}Sr/^{86}Sr$ ratio data for possible host rocks can be used to identify the location of the aquifer.

IV. CONCLUSIONS AND RECOMMENDATIONS

- Geochemical and isotope compositions of water ($\delta^{18}O$ and $\delta^{2}H$) are helpful tools in identifying the primeval nature of water in the STD SW reservoir:

  + The Miocene formation water is ancient seawater;

  + The Basement formation water is alternated fresh water.

- The $^{87}Sr/^{86}Sr$ isotope ratios of strontium dissolved in different kinds of water prove for the isotope equilibrium between formation water and sedimentary rock in Miocene reservoir as it was proposed earlier;

- The high Sr content (~ 8-10 mg/L) in the Basement water indicates the contribution of Sr from host rocks of the aquifer, so it is possible to identify aquifer location by the $^{87}Sr/^{86}Sr$ ratio analysis.

- Further application of the isotopic techniques in studies for the nature of formation water is recommended.

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