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Use of radium in studying water resources in Shanafiya-Samawa area- south Iraq

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Abstract

The water resources, Groundwater and surface water, in Shanafiya – Samawa area- southern Iraq were investigated using radium-226. The study examines the use of radium as tracer whether saline surface water (Sawa Lake) seeps and interacts with water of the Euphrates River and where groundwater interacts with surface water, Euphrates, Atshan River and Sawa Lake. As well as examine the radiological doses caused by consumption of these waters. Thirteen water samples were analysis for radium-226 content by precipitation with barium carrier by using gamma spectroscopy based on hyper-pure Germanium with efficiency 30%. The result shows that Sawa Lake contains radium concentration higher than that of groundwater and other sources of surface water. The high value of Ra in the Sawa Lake can be related to the high salinity. Groundwaters generally have Ra concentrations that broadly correlate with salinity or with total dissolved solids (TDS). The radium content in the different water depends on the hydrochemistry of water especially chlorine and TDS. The radium contents accumulate as clusters in different groups when plotted with chlorine and TDS which reveals no intermixing between the different water resources. While the relatively high concentration of Ra-226 in Sawa Lake may be caused by accumulation of Ra by maxing with groundwater according the hypotheses that states: in high salinity water system the dissolution of radium increases and accumulates in the solution. While low Ra-226 concentration in river water reveals that no intermixing has occurred between groundwater and river water. The annual effective doses due to ingestion of groundwater and river water in the study area were estimated to be less than the values recommended by IAEA and WHO.

Keywords: Radium, water resources, effective dose, Iraq.

استخدام نظير الراديوم في دراسة عن مصادر المياه في منطقة الشنافية – السماوة-جنوبي العراق

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الخلاصة

جرت دراسة مصادر المياه في منطقة الشنافية- السماوة جنوبي العراق باستخدام نظير الراديوم- 226 لمعرفة جدوى استخدام الراديوم-226 كعنصر اثر في حالة وجود تداخل مياه مالحة مثل مياه بحيرة ساوة مع المياه السطحية متمثلة في مياه نهر الفرات والعطشان واي تداخل بين المياه السطحية مع المياه الجوفية. كما تهدف الدراسة لبيان الجرعة الاشعاعية الناجمة عن استهلاك تلك المياه . تم تحليل اشعاعي لثلاثة عشر نمودجا لمياه منطقة الدراسة لتحديد تراكيز الراديوم-226 باستخدام منظومة تحليل اطياف كاما المستندة الى عداد

الجرمانيوم عالي النقاوة ذي الكفاءة 30%. بينت النتائج بان تراكيز الراديوم في مياه بحيرة ساوة اعلى منه في بقية مصادر المياه في المنطقة. وان القيم العالية نسبيا من الراديوم في مياه بحيرة ساوة مرتبط مع الملوحة العالية لمياه البحيرة. وان تراكيز الراديوم في المياه الجوفية تتناسب الى حد كبير مع محتوى الملوحة ومع محتوى المواد الصلبة الذائبة الكلية. كما بينت النتائج ان العلاقة بين محتوى الراديوم مع محتوى الكلورايد او المواد الصلبة الذائبة الكلية في المياه المختلفة تكون تجمعات بشكل مجاميع مختلفة تعكس عدم تداخل لتلك المياه المختلفة مع بعضها البعض. ان ارتفاع تراكيز الراديوم نسبيا في مياه بحيرة ساوة ربما تكون بسبب تجمع الراديوم نتيجة مزج المياه الجوفية في المنطقة مع مياه البحيرة كون ان ذوبان الراديوم في المياه ذات الملوحة العالية يزداد ويتجمع في المحلول. هذا ادى الى انتزاع الراديوم من المياه الجوفية وتجمعه بشكل ذائب في مياه بحيرة ساوة. بينما تدل تراكيز الراديوم الواطئة في مياه النهر الى عدم وجود تمازج مع المياه الجوفية او مع مياه البحيرة. جرى تحديد الجرغ المؤثرة السنوية الناجمة عن استهلاك مياه النهر او المياه الجوفية لاغراض الشرب في منطقة الدراسة وبينت النتائج ان قيم الجرغ كانت واطئة واقل من القيم الموصى بها من قبل الوكالة الدولية للطاقة الذرية او من قبل منظمة الصحة العالمية.

Introduction

There are four naturally radioactive isotopes of radium present in the environment. The most abundant among the naturally occurring isotopes are ^{226}Ra , an alpha emitter, with half-life of 1600 years [1]. The behaviour of Ra is similar to that of Ba due to the similarity of their ionic radii. In groundwater, radium is more likely to contain dissolved radium due to is introduced into natural water primarily through water-rock interactions. The major source for radium in surface water such as lakes and rivers is typically assumed to be derived from infiltrating groundwater.

The main radium sink in lakes is attributed to the adsorption to and the sedimentation of particulate matter. In rivers, radium adsorption can occur in the hyporheic zone as well as along streams, and short-lived radium isotopes can be generated in the bed sediments. Hence, the radium geochemistry in surface water is linked to the interactions between three possible components: groundwater, surface water and sediments [2].

The behavior of radium in groundwater was summarized by Porcelli and Swarzenski [2] as follows; Input by weathering, input by desorption, input by recoil from the aquifer matrix or from sorbed parent nuclides, input by decay from nuclides in solution, removal by adsorption, removal by decay, and removal by precipitation. In surface water, the major source for dissolved radium is infiltrating groundwater. Radium sinks in surface water are expected to vary with salinity. An additional potential sink for radium is co-precipitation with sulfate minerals. The amount of radium removed by co-precipitation would depend on the water chemistry and saturation state of different minerals.

The hypotheses for using radium as tracer that may be worked in this research are: in low saline surface water system representing by the Euphrates River and Atshan River, Ra is sequestered through adsorption onto suspended matter in the water if mixing occurs with groundwater which already contains higher radium, and where intermixing occurs between groundwater with high saline system (Sawa Lake) the radium sequestered in solution due to fact that dissolution of radium increase with salinity and accumulated in the solution and may be co-precipitation, with sulfate minerals, most notably BaSO_4 , occurred.[1,2]

The study area is located within Shanafiya (Diwania)-Samawa Area, SW -Iraq, within longitudes $44^\circ 33' 57''$ _ $45^\circ 15' 27''$ and latitudes $31^\circ 06'$ _ $31^\circ 36' 14''$. Figure (1) illustrates the study area including the locations of sampling stations

The current study examines the use of radium as tracer whether saline surface water (Sawa Lake) seeps and interacts with water of the Euphrates River and where groundwater interacts with surface water, Euphrates, Atshan River and Sawa Lake. As well as examine the radiological doses caused by consumption of these waters.

Geology of the study area

The study area and the surroundings are entirely covered by sedimentary rocks of the Cenozoic Era, ranging in age from Early Eocene up to Recent (Quaternary sediments). The Rus Formation of E. Eocene age consists of flaky yellowish marl with thin beds of nummulitic limestone and recrystallized limestone, partly silicified.[3]. Dammam Formation of is the only exposed formation of Paleogene

Epoch in the study area. It comprises limestone, dolomite, marl, and shale [4] with whitish grey porous dolomitized, locally chalky limestone. Dammam formation deposited in carbonate inner shelf lagoon and shoal[3]. It exposed in west and south west of the study area. The hydrogeological investigations in the Southern Desert [5] reflected that the Dammam Formation contains huge amounts of groundwater. This aquifer is considered as the main regional groundwater aquifer within the Southern Desert, due to wide extension and content of groundwater. The Euphrates formation is unconformably and extensively oversteps the Dammam Formation [6]. The Euphrates Formation is composed of shelly, chalky, well bedded, recrystallized, limestone [7]. The Early Miocene age was given to Euphrates Formation by [8].

The upper contact with Nfayel Formation is conformable based on the first appearance of Marl bed of Nfayel Formation about (2 m) in thickness [3, 7, and 9]. The gypsum of Fatha Formation is changed to limestone and marl toward the south, and this indicates that the basin is becoming deeper[10]. The given age of Nfayel formation is Middle Miocene age. Figure-2 shows geology of the study area and the surroundings.

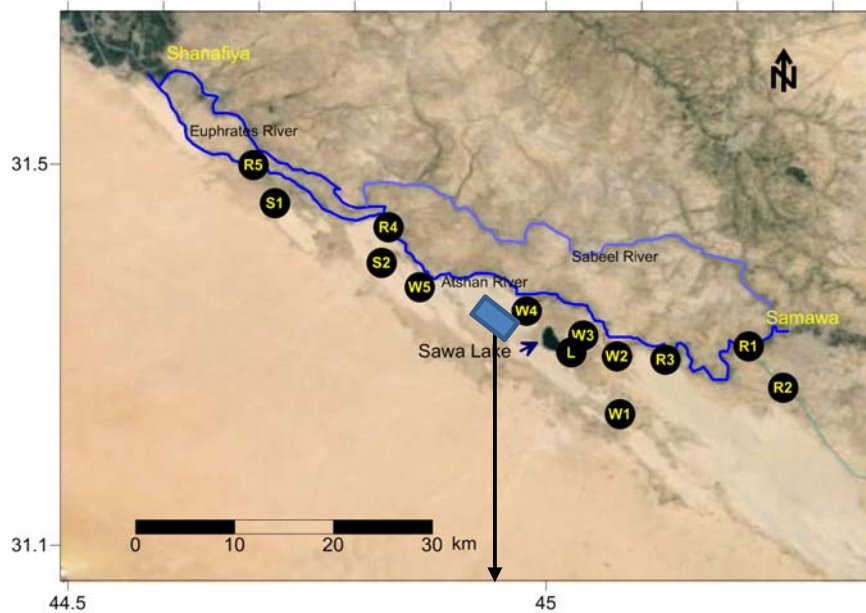
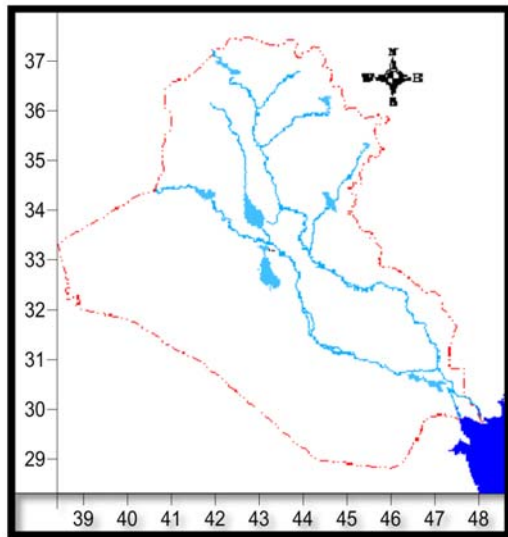


Figure 1-Map shows the study area and sampling stations.

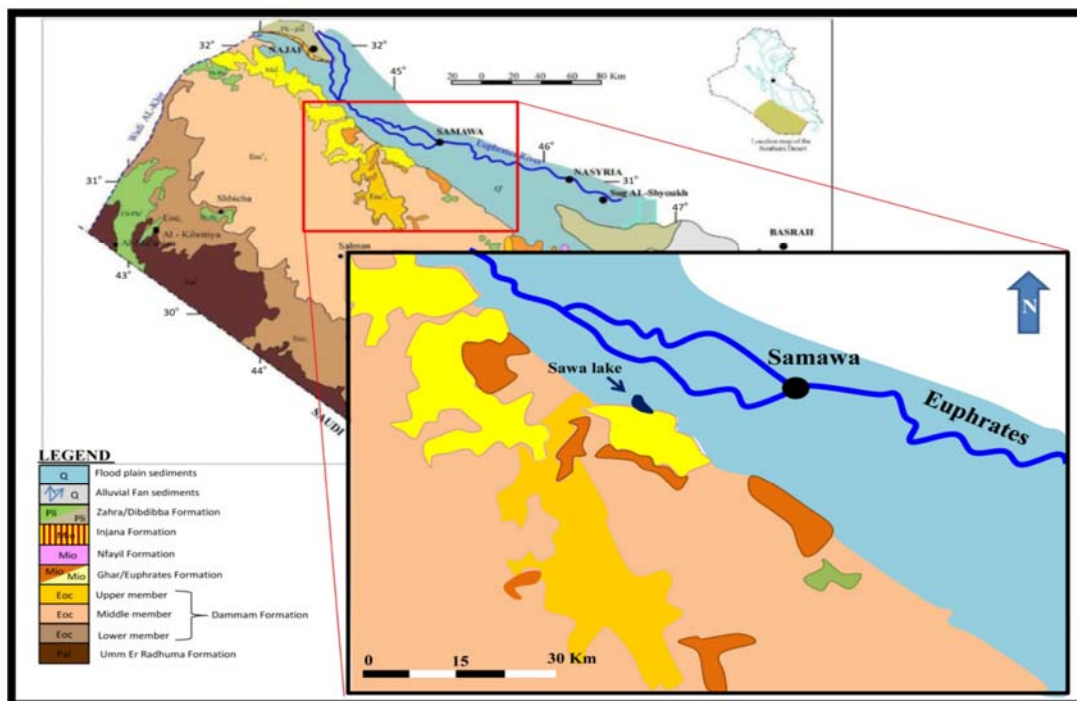


Figure 2- geological map of the study area and surroundings [11].

Materials and Methods

Radium-226 was measured in (13) water samples; (5) river water, (5) groundwater, (2) springs and (1) samples from Sawa Lake for two stages (figure- 1); the first was in October /2013(dry season) and the second one was in March /2014 (wet season). Preparing samples for radium analysis as follows [12]: 40 L of water sample is selected. To avoid adsorption and algae phenomena must be acidified to pH 1 with acid nitric (HNO_3 68%). Add 1 ml of H_2SO_4 per 10 liters of specimen. Pour drop wise 50 ml of the solution coach $\text{Ba}^{2+} 10 \text{ g l}^{-1}$ vigorously stirring the solution for analysis. Add 20 ml concentrated H_2SO_4 (98 %) to precipitate the sulphate. Decant the precipitate of barium sulfate $\text{Ba}(\text{Ra}) \text{SO}_4$ for about 24 h. Remove most of the supernatant with the aid of the pump and the rest to centrifuge about 2000 min^{-1} for 5 min to recover the precipitate .Wash the precipitate with nitric acid (18 %) in the centrifuge tube, shake and centrifuge . Rinse the precipitate with water twice under stirring and centrifuging for each operation. Dry the precipitate in the centrifuge tube in an oven at about 105°C for at least 24 h. Transfer the precipitate in a desiccator. Weigh the precipitate and packaged in a sealed container to avoid any loss of radon. Measure the content by gamma spectrometry. More details in IAEA [13] and Medley [14].Radium analysis were done in 2014 using gamma spectroscopy system in laboratories of Radiation Protection Center/Ministry of Environment - Iraq and the Jordanian Atomic Energy Commission. The hydrochemical analysis includes TDS and cations, anions were achieved in laboratories of Ministry of Science and Technology.

Results and Discussion

Radium-226 was measured in (13) water samples. The limitations of the small data set make it difficult to compare activity concentrations between ground and surface waters. However, this data set shows that the highest levels were found in Sawa Lake and ground waters while the lowest levels were found in surface waters.(Table -1)

The results of analysis for Ra-226 in Bq/l in the water resources in the study area indicate that ^{226}Ra activities in all water resources range between 4 mBq/l and 371 mBq/l and are consequently within the WHO drinking water regulations in 2008 of maximal 1000 mBq/l [15].

The average ^{226}Ra -activities in water samples are 0.00798, 0.0265, 0.114 and 0.371 Bq/l for River, wells, springs and Sawa Lake respectively. On the other hand, for contrast, the average activity of ^{226}Ra in sediments of Euphrates River and springs are 83.5 and 80.5 Bq/kg dry sediment respectively

[16] while the average activity of ^{226}Ra in the sediments of Sawa Lake is 27.8 Bq/kg dry sediment [17].

Table 1- The minimum, maximum and average concentrations of Ra-226, Cl and TDS in water samples in the study area.

Water resource	Radium concentration (Bq/l)			Cl (ppm)			TDS (ppm)		
	Min	Max	Average	Min	Max	Average	Min	Max	Average
River	0.00408	0.0168	0.00798	260	350	304	1951	2542	2221.2
Wells	0.012	0.045	0.0265	1599	2613	2025.3	4805	7982	6061
Springs	0.1001	0.1287	0.114	1234	1250	1242	3485	4390	3937.5
Sawa Lake	0.350	0.392	0.371	9056	9456	9256	2359	23660	23627.5

The activities of ^{226}Ra are linearly correlated with chloride and TDS (Figure -3 a and b). Groundwaters generally have Ra concentrations that broadly correlate with salinity or total dissolved solids (TDS) [18]. While Ra mobilization from rocks seems to be controlled by recoil, which is a physical process, Ra adsorption depends on the water chemistry and decreases with salinity, acidity, temperature, and reducing conditions [2, 19]). Consequently, saline [20, 21], reduced [22], acidic [23], and thermal [24] waters have typically high levels of Ra activities. Figure (3) shows that the activity of radium in Sawa Lake is enriched compared to the radium activity in the water of the wells, springs and river water.

The high value of Ra in the Sawa Lake can be related to the high salinity. As it is clearly shown (figure -3) that the concentration of Ra-226 in the different water resources clusters in different groups representing the different types of water resources, depending on the hydrochemistry, especially, on the salinity of the water and may reveals the no intermixing occurs between those resources. On the other hands, the relatively increase of radium in R3 station may be due to the influence of recharge from direct water flow from the spring and wells through the irrigation streams in this area to the river (field observations). In current study the concentration of Ra in groundwater is higher than that of river water which that means no mixing occurs between them.

Radium in surface water has low and narrow range of radium concentration in contrast to relatively high and wide range of radium concentration in groundwater [25-27]. The radium concentration in the Euphrates River is comparable with the concentration of radium in most other rivers in the world (Table -2.) although there are some of those rivers, such in China, Egypt, Germany and India, have higher concentration than that of the Euphrates River, which depend on geology of the bed rock and the potential contamination from artificial sources.

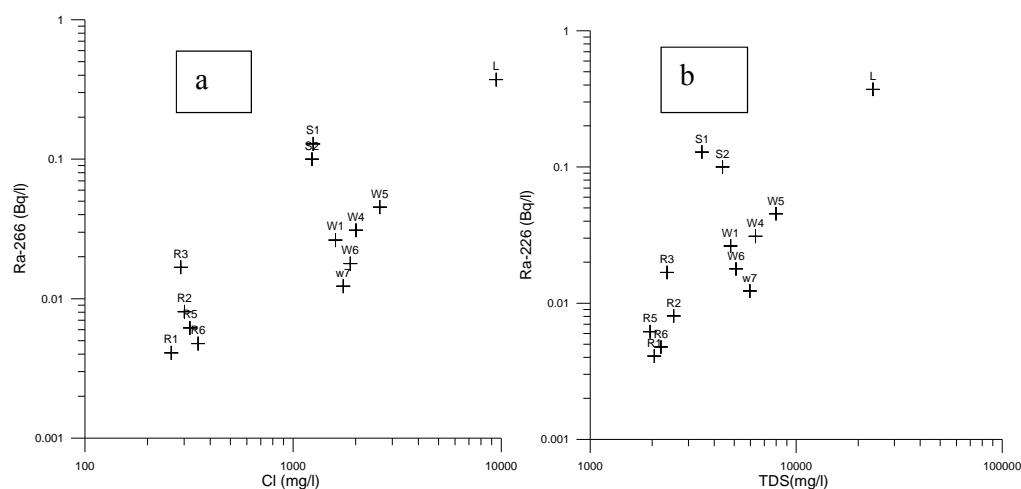


Figure 3- The relationship between (a) ^{226}Ra and Cl and (b) ^{226}Ra and TDS.

The Sawa Lake and springs have higher concentration than other surface water, because of the higher salinity they have. Radium is less likely to be adsorbed on particles as the salinity increase [20, 28-30] it is presumed that, under high salinity conditions, radium is conservative (i.e., its only sink is

radioactive decay). Raanan et al., [31] showed that low salinity surface water environments provide a significant sink for radium through adsorption to suspended matter. In current study the Sawa Lake, with higher salinity, conserves the radium that accumulated from recharge of groundwater leading to have higher concentration of radium than groundwater. Thus, in conclusion, that the source of water for wells may be connected to that of Sawa Lake or it may be the same source.

Another important source for radium nuclide in aquifers is alpha recoil [32]. Each alpha decay occurs with sufficient energy that the resulting daughter nuclide is propelled in a random direction for a distance of ~20 nm in most U/Th-bearing minerals. When such a daughter atom crosses the mineral-water interface, it can either stop within the groundwater or within another phase from where it can migrate back out through the damaged lattice. This is a purely physical process that is unaffected by groundwater chemistry

As stated earlier, Ra-226 in groundwater is derived mainly from interaction between the groundwater aquifer and radium-bearing minerals which are responsible for enhanced ^{226}Ra concentrations. Several studies have shown an increase in dissolved radium associated with increasing salinity, suggesting that radium is adsorbed from adsorption sites [20, 28-30, 33]. Other studies have shown that Ra adsorption generally decreases with acidity, temperature and reducing conditions [1]

Table 2- The radium concentration (mBq/l) in different rivers in the world [1] and radium concentration in the Euphrates River in the current study.

country	Ra-226	Reference
Australia: Magela Creek	18.5	[25]
Brazil: Amazon River	0.78–1.22	[25, 34]
Canada	4.07–13.7	[25, 35]
China	7.4–18.5	[25, 28]
Former Czechoslovakia	3.7–292	[25, 36]
Egypt: Nile River	3.7–155	[25, 37]
Germany	2.59–31	[25, 38]
India: Ganges River	20–48	[25, 39]
India: Ganges River	3.33	[25, 40]
United Kingdom: River Thames	18.5	[25, 41]
USA: Mississippi River	0.37–40	[25, 42]
USA: Hudson River	1.17	[25, 43]
USA: Suwannee River	7.4	[25, 27]
Japan (ten rivers)	1.48-5.18	[44]
Iraq- Euphrates River	7.98	Current Study

The concentration of radium in solution is consequently more closely related to the groundwater chemistry than to the radium concentration in the host rocks [1,2, 45]. High radium concentrations in groundwater may be found with rocks containing little uranium but in contact with saline water, whereas uraniumiferous rocks in contact with low salinity water might yield little radium [45].

Radiological Aspect

Dosimetric models to estimate human internal exposures subsequent to incorporation of radionuclides via ingestion have been developed by the ICRP; the most recent models for Ra isotopes are described in [46] for ingestion.

When analyzing the total annual effective dose to the human population from natural sources, the dose received by ingestion of long-lived natural radionuclides must be considered.

Annual effective doses (AED) per person resulting from the intake of the radionuclide ^{226}Ra through water in the study area were calculated by using the following equation:

$$\text{AED} = \sum I_i D_i$$

Where I_i is the annual intakes of radionuclide I (Bq y^{-1}) and the ingestion dose coefficient D_i for ^{226}Ra is $2.8 \times 10^{-7} \text{ SvBq}^{-1}$ [45]. The ingestion dose coefficients (Sv Bq^{-1}) reported by the International Commission on Radiological Protection (ICRP) [45] and IAEA [1] were used.

The computed annual effective dose received by the population according to this research (Table 3) is less than 1 mSv/y recommended by ICRP [46]. Moreover it is less than the effective dose equal to

0.1mSv/y which recommended in by WHO [15]. These values are very low compared with the annual dose limit of 2.4 mSv/y from the exposure of the public to all sources of radiation [47].

Table 3- The R-226 concentration, annual intake and the annual effective dose by ingestion of different water in the study area

ID	Ra-226 (Bq/l)	Annual intake (Bq/y)	effective dose(mSv/y)
R	0.00798	5.8254	1.63111E-03
W	0.0265	19.345	5.4166E-03
S	0.114	83.22	2.33016E-02

Conclusion

- The average ²²⁶Ra-activities in water samples are 0.00798, 0.0265, 0.114 and 0.371 Bq/l for river, wells, springs and Sawa Lake respectively.
- Groundwaters generally have Ra concentrations that broadly correlate with salinity or total dissolved solids (TDS).
- The activity of radium in Sawa Lake is enriched compared to the radium activity in the water of the wells, springs and river water. The high value of Ra in the Sawa Lake can be related to the high salinity.
- When the content of Ra-226 is plotted against Cl and TDS in the different water resources it gives clusters in different groups representing the different types of water resources. These clusters may be reveal the no intermixing occurs between deep groundwater with that of the river.
- The relatively high concentration of Ra-226 in Sawa Lake may be caused by accumulation of Ra by maxing with groundwater according the hypotheses that states: in high salinity water system the dissolution of radium increases and accumulates in the solution. That means water of Sawa Lake extracts radium from groundwater.
- While low Ra-226 concentration in river water reveals that no intermixing has occurred between groundwater and river water.
- For radio-ecological aspect, the annual effective radiation doses that caused by ingestion of the different water is very low and could be negligible.
- The use radium isotope in this study confirms that it is very important tool to investigate the hydrological system and water management.

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