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^{228}Ra/^{226}Ra activity ratio in groundwater around Mount Fuji, Japan

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Abstract

We estimated the groundwater age from ^{228}Ra/^{226}Ra ratios in young groundwater and relevant rocks in the volcanic area of the Kakitagawa River around Mount Fuji, Japan, and compared our results with those from ^{3}H/^{3}He age determination. The groundwater residence time estimated from the ^{228}Ra/^{226}Ra activity ratio in groundwater and relevant rocks agreed well with the ^{3}H/^{3}He age, suggesting that the ^{228}Ra/^{226}Ra ratio of groundwater can be used to estimate residence time of young groundwater in volcanic areas.

1. Introduction

Groundwater younger than 100 years can be dated from environmental radioactivity of isotopes with short half-lives (^{3}H/^{3}He [1, 2], ^{85}Kr [3, 4]), or chemicals like CFC-12 [4]. The ^{3}H/^{3}He dating method accurately estimates the residence time of shallow groundwater. One of the advantages of this

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method is small sample volume: only several tens of milliliters of groundwater. The ^{3}He in groundwater originates from 3 sources: tritiogenic He, mantle He, and radiogenic He produced in the rock. Especially as the contribution of mantle He is greater than those from radiogenic and tritiogenic sources, when groundwater is dated with ^{3}H/^{3}He in volcanic areas, we have to determine the ratio of the three sources of ^{3}He. In contrast, as ^{85}Kr originates only from the atmosphere, it is an excellent tracer for dating groundwater in volcanic areas. However, as the concentration of ^{85}Kr in groundwater is ultra low, for measurement,^{85}Kr must be separated from about 10^{4} l of ground water.

Groundwater dating by these methods has both advantages and disadvantages, but the disadvantages of the individual methods can be offset by using multiple tracers. Therefore, development of additional groundwater dating techniques is desired.

Ra-228 and Ra-226 are progenies of Th and U, respectively. The ^{228}Ra/^{226}Ra ratio in groundwater depends on the Th/U in the relevant rocks. As the ^{228}Ra and ^{226}Ra in shallow groundwater in volcanic areas originates only from rock, and collecting radium isotopes from groundwater is much easier than collecting ^{85}Kr, Ra may be a good tracer of groundwater age in volcanic areas. In this study, we evaluated the application of radium isotopes to groundwater dating in a volcanic area. As the ^{228}Ra half-life is short (5.8 y) and the ^{226}Ra half-life is long (1600 y), the ^{228}Ra/^{226}Ra ratio in natural waters is used to trace water movement [5–7] or estimate water origin [8]. We estimated groundwater age from ^{228}Ra/^{226}Ra ratios in groundwater and relevant rocks in a volcanic area. We compared the ^{228}Ra/^{226}Ra groundwater ages with those from ^{3}H/^{3}He dating for the Kakitagawa River near Mount Fuji, Japan. The water of the Kakitagawa River originates from Mount Fuji groundwater.

2. Experiments

2.1 Kakitagawa River observation area

The observation area is in the northeast of the Kanto Mountains. The Kakitagawa River is in the eastern district of Shizuoka Prefecture. Sampling points for ^{228}Ra/^{226}Ra ratio in the Kakitagawa River were located along a basaltic lava flow (Mishima lava flow) area of Mount Fuji (Figure 1). The geological detail for the Kakitagawa area is described by Tsuya [9]. The basaltic rock samples for Kakitagawa were collected from the Mishima lava flow.

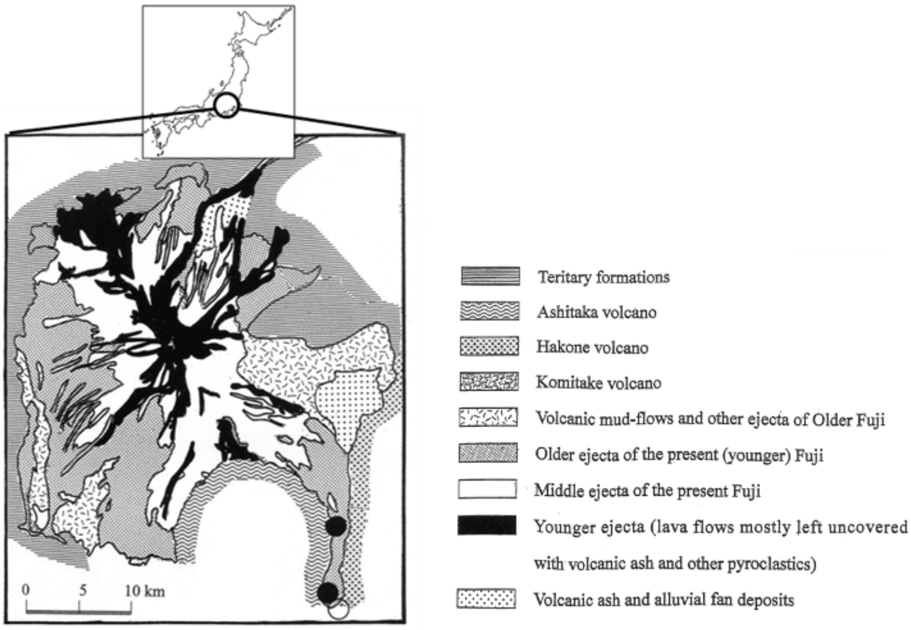

Figure 1: Sampling sites of Kakitagawa river and relevant rock. ●: rock, ○: groundwater.

2.2 Preparation of Mn-impregnated acrylic fiber and groundwater sampling

Mn-impregnated acrylic fiber was prepared after the procedure of Moore [10]: 100 g of acrylic fiber was put into 1 l of $0.3\,\mathrm{mol}\cdot\mathrm{l}^{-1}$ $KMnO_4$ solution and kept for 12 h at 50 °C to form Mn-impregnated acrylic fiber; the product was then rinsed well with flowing deionized water and dried completely in an electric oven at 50 °C. A detailed description on the Mn-impregnated acrylic fiber is given by Ohta *et al.* [11].

^{226}Ra and ^{228}Ra were collected from groundwater flowing through this Mn-impregnated acrylic fiber. Groundwater samples for measuring radium isotopes were collected three times a year from the Kakitagawa River from 2005 to 2006.

2.3 Investigation of (^{228}Ra/^{226}Ra activity ratio)$_{T=0}$

We measured the ratio (^{228}Ra/^{226}Ra)$_{T=0}$ with the following methods:

1) Leaching test from rock samples,

Table 1: The activity ratio of the supplied radium from rock sample to water. ^{228}Ra/^{226}Ra$_{\mathrm{(rock)}}$: activity ratio of ^{228}Ra/^{226}Ra in rock, ^{228}Ra/^{226}Ra$_{\mathrm{(water)}}$: activity ratio of ^{228}Ra/^{226}Ra in water supplied from rock sample, AR: activity ratio between water and rock of ^{228}Ra/^{226}Ra.

Rock type	^{228}Ra/^{226}Ra$_{\mathrm{(rock)}}$	^{228}Ra/^{226}Ra$_{\mathrm{(water)}}$	AR$_{\mathrm{(water)}}$/ AR$_{\mathrm{(rock)}}$
Granite	1.0 ± 0.1 ($n = 2$)	1.8 ± 0.4 ($n = 3$)	1.8 ± 0.4

2) ^{228}Ra/^{226}Ra activity ratio in mountain water (see below) and relevant rock.

Rock samples (granite) were sliced and placed in a vessel with stainless steel utensils. The vessel was filled with pure water that had been de-aerated with a vacuum pump. The samples were then stored with the water for about 1 month, after which the water was separated from rock and the radium isotopes were collected from the water onto 20 g of a cation exchange resin (H form).

Ra isotopes in mountain water were collected from the Okutama area in the western part of Tokyo and from the island of Izu-Oshima, Japan. Mn-impregnated acrylic fiber was put at the bottom of the cliff after rain, and radium isotopes supplied from rock with a minimal residence time to water were collected on the fiber.

2.4 Measurement of ^{228}Ra and ^{226}Ra

The Mn-impregnated acrylic fiber with the collected Ra isotopes was dried and packed into an air-tight tin canister (76 mm $\phi \times$ 24 mm H) for γ-ray spectrometry and stored for one month to allow ^{214}Pb and ^{228}Ac to reach radioactive equilibrium with ^{226}Ra and ^{228}Ra, respectively. Radioactivity of ^{214}Pb and ^{228}Ac was determined from the 351 keV and 911 keV γ-rays, respectively. Details of the methods for sample collection and determination of radioactivity are available elsewhere [12, 13].

3. Results and discussion

3.1 Investigation of (^{228}Ra/^{226}Ra)$_{T=0}$

Table 1 lists the average ^{228}Ra/^{226}Ra ratio in the granite and the water supplied from the rock samples, and the ^{228}Ra/^{226}Ra activity ratio of water to rock. The average ^{228}Ra/^{226}Ra activity ratios in the granite rock sample

Table 2: The activity ratio in mountain water. $^{228}Ra/^{226}Ra_{(rock)avg}$: average of $^{228}Ra/^{226}Ra$ in rock, $^{228}Ra/^{226}Ra_{(water)avg}$: average of $^{228}Ra/^{226}Ra$ in mountain water, AR: activity ratio between water and rock of $^{228}Ra/^{226}Ra$.

Geology	$^{228}Ra/^{226}Ra_{(rock)avg}$	$^{228}Ra/^{226}Ra_{(water)avg}$	$AR_{(water)}/AR_{(rock)}$
Basaltic rock area	0.5 ± 0.1	1.2 ± 0.3	2.4 ± 0.8
Sedimentary rock area*	1.5 ± 0.1	2.5 ± 0.3	1.7 ± 0.2
Sedimentary rock area*	1.5 ± 0.1	$2.8 \pm 0.0_4$	1.9 ± 0.2

* Detailed information is reported by Nakano-Ohta and Sato [14].

and water exposed to the rock samples were 1.0 ± 0.1 ($n = 2$) and 1.8 ± 0.4 ($n = 4$), respectively. The average $^{228}Ra/^{226}Ra$ ratio of the water exposed to the rock was 1.8 times higher than that of the relevant rock.

Table 2 shows the $^{228}Ra/^{226}Ra$ activity ratio of rock and mountain water from two sedimentary rock areas and a basaltic area. The average $^{228}Ra/^{226}Ra$ activity ratios in mountain water from two sedimentary rock areas of Okutama area were 2.5 and 2.8 ($n = 2$). The average activity ratio for the sedimentary rock from both of these areas was 1.5 ± 0.1 ($n = 3$). The ratio in water was 1.7 or 1.9 times those in the relevant sedimentary rock. In the basaltic area, Izu-Oshima, the $^{228}Ra/^{226}Ra$ activity ratio in the water and the rock were 1.2 and 0.5, respectively, and the $^{228}Ra/^{226}Ra$ ratio in the water was 2.4 ± 0.8 times that in the basaltic rock. As shown in Tables 1 and 2, the $^{228}Ra/^{226}Ra$ ratio of the water supplied from the relevant rocks was approximately double that of the rock samples. Thus, the $(^{228}Ra/^{226}Ra)_{T=0}$ in groundwater may be nearly double that in the relevant rock. Furthermore, the $^{228}Ra/^{226}Ra$ ratios in water and those in local rocks are almost the same for basaltic, sedimentary rock, and granite areas.

3.2 ^{228}Ra/^{226}Ra in the Kakitagawa River and the Mishima lava flow

Table 3 shows the $^{228}Ra/^{226}Ra$ activity ratio in groundwater of the Kakitagawa River and Mishima lava flow. The $^{228}Ra/^{226}Ra$ activity ratio of 4 basaltic lava-flows in the Kakitagawa area ranged from 0.8 to 1.0 (average

Table 3: The activity ratio in groundwater. ^{228}Ra/^{226}Ra$_{\text{(rock)avg}}$: average of ^{228}Ra/^{226}Ra in rock, ^{228}Ra/^{226}Ra$_{\text{(water)}}$: range of ^{228}Ra/^{226}Ra in Kakitagawa River water, AR: activity ratio of ^{228}Ra/^{226}Ra, ^{228}Ra/^{226}Ra$_{(T=0)}$: initial value of ^{228}Ra/^{226}Ra in water estimated from the rock.

	^{228}Ra/^{226}Ra$_{\text{(rock)avg}}$	^{228}Ra/^{226}Ra$_{\text{(water)}}$	^{228}Ra/^{226}Ra$_{(T=0)}$
Mishima lava flow	0.9 ± 0.1 ($n = 4$)	0.8-1.1 ($n = 3$)	1.8

value: 0.9 ± 0.1). Assuming that the Ra isotopes in groundwater were supplied from the overlying rock in the aquifer with constant ratio, the ^{228}Ra/^{226}Ra activity ratio may decrease with increase in the residence time of the groundwater in the aquifer. Supposing that the (^{228}Ra/^{226}Ra)$_{T=0}$ activity ratio in the groundwater is twice that of the relevant rocks, the activity ratio for the youngest groundwater of the Mishima lava flow may be 1.8, and (^{228}Ra/^{226}Ra)$_{T=0}$ may be assumed to be 1.8. Based on this assumption, from the initial ^{228}Ra/^{226}Ra activity ratio of groundwater (from 0.8 to 1.1), we estimate the residence time of the Kakitagawa groundwater to be from 9 to 16 years. In fact, the residence time of the groundwater of the Kakitagawa River was 10 years determined by ^{3}H/^{3}He groundwater dating [2]. The residence time of the Kakitagawa estimated by the present Ra in the groundwater and the relevant rock agrees well with that determined by ^{3}H/^{3}He groundwater dating, suggesting that the Ra isotope ratios of groundwater can be used to estimate the residence time of young groundwater in volcanic areas.

4. Summary and conclusions

The ^{228}Ra/^{226}Ra ratios in groundwater and rock samples suggest that:

(1) The average activity ratio of ^{228}Ra/^{226}Ra in water supplied from rock was about double that of the rock samples. Therefore, the (^{228}Ra/^{226}Ra)$_{T=0}$ ratio when the radium isotopes were supplied into groundwater from the relevant rock might have been nearly the double of that of the relevant rock.

(2) The activity ratio in the groundwater of the shortest residence time of Kakitagawa was estimated from the relevant rocks (Mishima lava flow) to be 1.8, resulting in a residence time of Kakitagawa river waters from 9 to 16 years. The residence time of Kakitagawa estimated from

the $^{228}Ra/^{226}Ra$ ratios agrees well with the $^{3}H/^{3}He$ age. Our results suggest that the $^{228}Ra/^{226}Ra$ activity ratio can be used to date young groundwater in volcanic areas.

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