Article

$^{228}\text{Ra}/^{226}\text{Ra}$ Activity Ratio in River Water in Okutama Area, Western District of Tokyo

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The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios obtained by the observations with 3 river waters, 2 hot-spring waters and 12 samples from sedimentary rock occurring in Okutama area, western district of Tokyo, ranged from 1.0 to 2.9, from 0.7 to 0.8 and from 1.2 to 1.7, respectively. The average values were $2.0 \pm 0.0_3$ for river waters and 1.4 ± 0.1 for sedimentary rocks, suggesting that the activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ in river water tends to be larger than that in the associated sedimentary rocks. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in hot-spring waters staying in the deeper aquifer with longer residence time may tend to be smaller than those in river waters with shorter residence time. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in river waters showed that the activity ratios tended to decrease after heavier rainfalls than 20 mm/d. The decrease in the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in river waters suggested that heavy rainfalls forced the deeper underground water with long residence time to discharge into river water.

Key Words : radium-226, radium-228, activity ratio, river water, hot-spring water,
Okutama area

1. Introduction

Concentration level of Ra isotopes in river waters is $\text{nBq} \cdot \text{L}^{-1}$ or less in the order of magnitude^{(1)–(4)}. As ^{226}Ra and ^{228}Ra are progenies of ^{238}U and ^{232}Th , respectively, the activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ in water is essentially dependent on the elemental composition ratio of Th/U in the rock with which the water is in contact. Wide range of $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters, spring waters and underground waters has been observed by many workers^{(1)–(11)}. Sturchio et al.⁽⁵⁾ reported that $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios of 0.06–1.48 in underground waters in Paleozoic carbonate aquifer, mid-continent, USA, was similar to those in the rocks of 0.21–1.53

within the aquifer. Zhuo et al.⁽⁶⁾ reported that the activity ratio in the underground water in Fujian Province, China, ranged from 0.3 to 4.1, and that concentrations of ^{222}Rn in underground waters from granitic aquifer was higher than that from the aquifer of metamorphic rocks and sedimentary rocks, suggesting that concentrations of ^{222}Rn in underground water varied with the rock type of the constituent of the aquifers. The activity ratios in hot-spring waters in Japan were reported to range from 0.05 to $18^{(7)–(9)}$.

The activity ratios of $^{228}\text{Ra}/^{226}\text{Ra}$ in the river waters may also provide informations on the mixing of waters from different origins. The activity ratio in Mississippi river water at several observation points located along the valley of 400 km between St. Francisville and Venice,

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Fig. 1 Geological map (after Ohmori¹⁴), modified) around sampling points of hot-spring waters, river waters and relevant rocks samples. ● : Hot-spring water, ○ : River water, ■ : Sedimentary rock

Louisiana, ranged from 0.1 to 1.1¹⁾. The observation in Morocco showed that the averaged $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios were 1.1 for river waters, 0.13 for spring waters, 0.70 for well waters and 0.3 for cold spring waters²⁾. The activity ratio in shallow underground waters, river waters and relevant rocks of the upper Rhine valley in France ranged from 2.1 to 2.2, from 0.7 to 2.0 and from 0.4 to 1.4, respectively³⁾. The activity ratio in underground waters and river waters in Okinawa, Japan, ranged from 0.5 to 1.3⁴⁾.

Hakam et al.²⁾ made a distribution map of radionuclides, ^{238}U , ^{226}Ra and ^{228}Ra , in natural waters of Morocco, which was helpful to distinguish the origin of particular radioactivity: pollution or natural origin. Eikenberg et al.³⁾ reported that $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios and $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios together with concentrations of Ra and Sr could be used as a geochemical tracer to distinguish types of waters and their interactions.

This report deals with $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios observed with river waters in western district of Tokyo together with the associated hot-spring waters and the relevant sedimentary rocks: 69 observations with 3 river waters, with 2 hot-spring waters and with 12 sedimentary rock samples.

2. Experiments

2.1 Observation area

Observation area is located in the southeastern part of Kanto Mountains. The geology belongs to Chichibu and Shimanto formations of the outer zone of Southwest Japan. The geological details are described by Sakai¹²⁾ and Mitsuchi¹³⁾. Sampling sites of river waters, hot-spring waters and sedimentary rocks are shown in Fig. 1.

Two hot-springs, Tsuru-no-yu and Nomekoi-no-yu, are located in Shimanto formation area^{15)–18)}. The chemical characteristics of the

Table 1 $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in sedimentary rocks samples

Geology	Rock type	$^{228}\text{Ra}/^{226}\text{Ra}$
Chichibu formation	Sandstone and shale	1.3 ± 0.1
	Sandstone and shale	1.5 ± 0.1
	Chart, siliceous shale and alternated bed of sandstone and shale	1.6 ± 0.1
Average		1.5 ± 0.1
Shimanto formation	Sandstone and crayslate	$1.3 \pm 0.0_4$
	Sandstone and clayslate	1.7 ± 0.1
	Clayslate and sandstone	1.4 ± 0.1
	Clayslate and sandstone	1.2 ± 0.1
	Clayslate and sandstone	1.4 ± 0.4
	Clayslate and sandstone	1.4 ± 0.1
	Clayslate and sandstone	1.3 ± 0.2
	Clayslate and sandstone	1.2 ± 0.1
	Clayslate and sandstone	1.2 ± 0.3
Average		1.3 ± 0.1

two hot-spring waters belong to alkaline simple thermal spring containing hydrogen sulfide^{15), 17)}.

Tsuru-no-yu hot-spring water is located below the dam of Okutama-ko lake. Kanroji et al.¹⁵⁾ reported that the water level of the hot-spring water is closely related to that of the Okutama-ko lake, and the hot-spring water is supposed to be partly fed by the lake water. Nomekoi-no-yu hot-spring water is known to change in color and in smell every day, implying mixing of waters from different sources with the hot-spring water. The hot-spring water of Tsuru-no-yu is welled up from the depth of 300 m¹⁵⁾ and Nomekoi-no-yu from the depth of 1 100 – 1 500 m¹⁹⁾.

Observation points of $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters of Nippara-gawa, Tamagawa and Asakawa river waters were also located in Chichibu and Shimanto formation areas.

The sedimentary rock samples were also collected from Chichibu and Shimanto formations.

2·2 Collection of Ra isotopes

The Ra isotopes in the two hot-spring waters were collected from 80 L of water samples by 35 g of cation exchange resin (Dowex 50 X8, #100 – 200, in H form prepared by $2 \text{ mol} \cdot \text{L}^{-1}$ HCl) in a column ($9.6 \text{ cm}^2 \times 7.5 \text{ cm}$). The Ra isotopes in river waters were collected by 60 g of Mn-impregnated acrylic fibers, collecting material, by placing in the stream of the river waters for several days. The Mn-impregnated acrylic fiber was prepared after the procedure given by Moore²⁰⁾: a 100 g of acrylic fiber was soaked in a 1 L of $0.3 \text{ mol} \cdot \text{L}^{-1}$ solution of KMnO_4 for 12 h at 50°C to form Mn-impregnated acrylic fiber, and the product was rinsed well with running deionized water and dried completely in an electric drier oven at 50°C . Detailed description on the Mn-impregnated acrylic fiber is given by Ohta et al²¹⁾.

2·3 Measurement of Ra isotopes

The cation exchange resin and the Mn-im-

Table 2 $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters

River waters	Sampling date			$^{228}\text{Ra}/^{226}\text{Ra}$
Nippara-gawa river	Dec 2	- Dec 3,	2001	1.3 ± 0.3
	Feb 5	- Feb 5,	2002	2.2 ± 0.3
	March 24	- March 24,	2002	1.5 ± 0.4
	Jan 1	- Jan 3,	2003	1.4 ± 0.1
	Jan 3	- Jan 8,	2003	1.4 ± 0.1
	Jan 8	- Jan 14,	2003	1.4 ± 0.1
	Jan 14	- Jan 21,	2003	$1.5 \pm 0.0_4$
	Jan 21	- Jan 28,	2003	$1.4 \pm 0.0_4$
	March 22	- March 22,	2003	$1.9 \pm 0.0_4$
	April 8	- April 9,	2003	1.5 ± 0.1
	May 7	- May 9,	2003	1.3 ± 0.2
	May 20	- May 23,	2003	1.3 ± 0.1
	June 3	- June 6,	2003	1.5 ± 0.1
	June 21	- June 24,	2003	1.5 ± 0.1
	July 5	- July 8,	2003	1.5 ± 0.1
	July 18	- July 21,	2003	1.5 ± 0.1
	Oct 28	- Oct 30,	2003	1.2 ± 0.1
	Dec 3	- Dec 6,	2003	1.3 ± 0.1
	Dec 17	- Dec 20,	2003	1.0 ± 0.1
Average				$1.5 \pm 0.0_4$
Tamagawa river				
Osawa	July 13	- July 13,	2002	1.8 ± 0.8
Mizune-sawa	July 13	- July 13,	2002	2.4 ± 0.9
Una-sawa	July 13	- July 13,	2002	1.6 ± 0.3
Konaka-sawa	March 24	- March 24,	2002	1.1 ± 0.3
Average				1.7 ± 0.3
Asakawa river	Jan 13	- Jan 13,	2002	1.7 ± 0.4
	March 31	- March 31,	2002	1.4 ± 0.5
	June 16	- June 16,	2002	1.6 ± 0.3
	April 4	- April 8,	2003	1.6 ± 0.3
	April 18	- April 22,	2003	2.0 ± 0.1
	May 3	- May 6,	2003	2.6 ± 0.6
	May 17	- May 20,	2003	1.9 ± 0.3
	June 1	- June 4,	2003	1.7 ± 0.2
	June 14	- June 17,	2003	2.1 ± 0.3
	June 28	- July 1,	2003	2.2 ± 0.2
	July 13	- July 16,	2003	1.9 ± 0.2
	July 26	- July 29,	2003	$1.8 \pm 0.0_4$
	Aug 26	- Aug 29,	2003	1.1 ± 0.1
	Sept 6	- Sept 9,	2003	2.1 ± 0.2
	Sept 18	- Sept 22,	2003	1.8 ± 0.1
	Oct 2	- Oct 7,	2003	2.1 ± 0.1
	Oct 14	- Oct 18,	2003	1.7 ± 0.1
	Nov 9	- Nov 13,	2003	2.1 ± 0.2
	Dec 6	- Dec 9,	2003	2.1 ± 0.4
	Dec 19	- Dec 22,	2003	1.9 ± 0.1
	Dec 22	- Dec 25,	2003	2.1 ± 0.1
	Dec 25	- Dec 28,	2003	2.5 ± 0.5
	Dec 28	- Dec 31,	2003	2.7 ± 0.3
	Dec 31	- Jan 3,	2004	2.5 ± 0.1
	Jan 6	- Jan 9,	2004	2.9 ± 0.3

River waters	Sampling date			$^{228}\text{Ra}/^{226}\text{Ra}$
Asakawa river	May 1	- May 3,	2004	2.1 ± 0.1
	May 3	- May 4,	2004	2.2 ± 0.1
	May 4	- May 7,	2004	2.2 ± 0.1
	May 7	- May 10,	2004	2.0 ± 0.1
	May 10	- May 13,	2004	2.2 ± 0.1
	May 13	- May 16,	2004	1.9 ± 0.1
	May 16	- May 19,	2004	2.4 ± 0.4
	May 19	- May 22,	2004	1.9 ± 0.1
	May 22	- May 25,	2004	2.0 ± 0.1
	May 25	- May 28,	2004	2.3 ± 0.3
	May 28	- May 30,	2004	2.4 ± 0.3
	May 30	- Jun. 3,	2004	2.0 ± 0.2
	June 3	- June 6,	2004	2.5 ± 0.1
	June 6	- June 9,	2004	2.2 ± 0.1
	June 9	- June 12,	2004	1.9 ± 0.1
	June 12	- June 15,	2004	1.6 ± 0.1
	June 15	- June 18,	2004	2.1 ± 0.1
	June 21	- June 24,	2004	2.6 ± 0.2
	June 24	- June 27,	2004	1.9 ± 0.2
	June 27	- June 30,	2004	1.8 ± 0.2
	June 30	- July 3,	2004	2.5 ± 0.3
Average				$2.1 \pm 0.0_4$

tive γ -ray spectrometry. The samples for the γ -ray measurement were stored for 1 month in order that ^{214}Pb and ^{228}Ac reached the radioactive equilibrium with ^{226}Ra and ^{228}Ra , respectively. Gamma-ray intensities were measured with an HPGe γ -ray spectrometer. Radioactivity of ^{214}Pb and ^{228}Ac were determined by the 351 keV and the 911 keV γ -rays, respectively. Detailed discussion for the present spectrometry is described by Saito et al⁸⁾.

3. Results and Discussion

3.1 $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in rock samples, river waters and hot-spring waters

Table 1 gives the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios observed with 12 sedimentary rock samples from Chichibu and Shimanto formations. The activity ratios range from 1.2 to 1.7 for Chichibu and Shimanto formations with averaged values of 1.5 ± 0.1 for Chichibu formation and 1.3 ± 0.1 for Shimanto formation, respectively. Table 2 lists the observed $^{228}\text{Ra}/^{226}\text{Ra}$ ac-

pregnated acrylic fiber recovered were dried to pack into an air-tight canister of tin-can (67 mm $\phi \times$ 15 mm H or 76 mm $\phi \times$ 24 mm H) for γ -ray spectrometry. A 10 – 60 g of rock samples were crushed into a grain size of 20 – 50 mesh and packed into the canisters for non-destructive

Table 3 $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in hot-spring waters

Hot-spring waters	Sampling date	$^{228}\text{Ra}/^{226}\text{Ra}$
Nomekoi-no-yu	May 14, 2003	0.8 ± 0.3
Tsuru-no-yu	May 20, 2003	0.7 ± 0.3

Fig. 2 Histogram for $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters, relevant rocks and hot-spring waters.

tivity ratios in 3 river waters of Nippara-gawa, Tamagawa and Asakawa rivers. Activity ratios scattered widely from 1.0 to 2.9 with the averaged values of 1.5 ± 0.0 , 1.7 ± 0.3 and 2.1 ± 0.0 , respectively. The averaged values are $2.0 \pm 0.0_3$ for river waters and 1.4 ± 0.1 for the relevant rock samples: the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters tended to be larger than that in the relevant rocks. The activity ratios for the 2 hot-spring waters are 0.7 ± 0.3 and 0.8 ± 0.3 as given in Table 3. The activity ratios are apparently smaller than those of river waters and relevant rock samples. The distributions of the observed $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters, in the relevant rock samples and in the hot-spring waters are compiled in Fig. 2.

Observation was also made with the activity

ratios of sinter deposits from a hot-spring water in southern Kyushu. The activity ratios were 5.8 ± 0.1 and 3.2 ± 0.1 , while those of the relevant rock samples from the geological layers from which the hot-spring waters were inferred to be gathered ranged from 1.4 to 1.6 with the averaged value of 1.6 ± 0.1 as given in Table 4.

However, the activity ratios in shallow underground water and in river waters in the upper Rhine valley was similar to those in the leachates of the bottom sediment of the river³⁾.

The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios of 0.06 – 1.48 in the underground water in Paleozoic carbonate aquifer, mid-continent, USA, is also similar to those of 0.21 – 1.53 in the aquifer rocks⁵⁾.

Even though Ra isotopes in natural waters may be the product mainly by chemical leaching from the source rock, a small amount of Ra isotopes produced by the α -recoil ejection from the parent Th isotopes can be added. As the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of Ra isotopes immediately after the emission by the α -recoil process is calculated to be 276 times larger than that in the original material, $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in the water tends to be larger than that in the rock in contact.

3.2 $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios and residence time of underground water

When the amount of Ra isotopes supplied into water from a unit surface of a rock in a unit time by chemical leaching and α -recoil process is expressed by $N_0(\text{Ra}) (\text{sec}^{-1} \cdot \text{cm}^{-2})$,

Table 4 Concentrations of ^{228}Ra , ^{226}Ra and $^{228}\text{Ra}/^{226}\text{Ra}$ in sinter deposits from Kyomachi hot-spring water and in relevant rocks

Hot-spring	$^{228}\text{Ra} / \text{mBq} \cdot \text{g}^{-1}$	$^{226}\text{Ra} / \text{mBq} \cdot \text{g}^{-1}$	$^{228}\text{Ra}/^{226}\text{Ra}$
Kyomachi hot-spring (1)	855 ± 10	148 ± 2	5.8 ± 0.1
(2)	351 ± 3	110 ± 1	3.2 ± 0.1
Average			4.5 ± 0.1
Geology			
Kakuto Basin propylite (1)	23 ± 4	14 ± 2	1.6 ± 0.3
(2)	50 ± 10	31 ± 2	1.6 ± 0.3
(3)	34 ± 4	25 ± 1	1.4 ± 0.1
Ikemure formation	37 ± 6	27 ± 4	1.4 ± 0.1
Kyomachi formation	50 ± 6	32 ± 5	1.6 ± 0.3
Ito pyroclastic flow	56 ± 12	29 ± 5	1.9 ± 0.4
Average			1.6 ± 0.1

Fig. 3 Variation in $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in waters with residence time.

the total amount of the Ra isotopes accumulated in the water $N(\text{Ra})$ after the time T will be

$$N(\text{Ra}) = N_0(\text{Ra}) [1 - \exp(-\lambda(\text{Ra})T)] / \lambda(\text{Ra})$$

where $\lambda(\text{Ra})$ represents decay constant of the radium isotope. The isotopic ratio of the Ra isotopes will be

$$\frac{N(^{228}\text{Ra})}{N(^{226}\text{Ra})} = \frac{N_0(^{228}\text{Ra}) [1 - \exp(-\lambda(^{228}\text{Ra})T)] / \lambda(^{228}\text{Ra})}{N_0(^{226}\text{Ra}) [1 - \exp(-\lambda(^{226}\text{Ra})T)] / \lambda(^{226}\text{Ra})}$$

As the largest value of the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in the present river waters so far ob-

served was 3, the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in the youngest ($T \approx 0$) ground water can be assumed to be 3, which corresponds to the isotopic ratio of $N_0(^{228}\text{Ra})/N_0(^{226}\text{Ra}) = 0.011$. The $^{228}\text{Ra}/^{226}\text{Ra}$ isotopic ratio will be

$$\frac{N_0(^{228}\text{Ra})}{N_0(^{226}\text{Ra})} = 0.011 \frac{[1 - \exp(-\lambda(^{228}\text{Ra})T)] / \lambda(^{228}\text{Ra})}{[1 - \exp(-\lambda(^{226}\text{Ra})T)] / \lambda(^{226}\text{Ra})}$$

or the activity ratio $A(^{228}\text{Ra})/A(^{226}\text{Ra})$ will be

$$\begin{aligned} \frac{A(^{228}\text{Ra})}{A(^{226}\text{Ra})} &= \frac{A_0(^{228}\text{Ra}) [1 - \exp(-\lambda(^{228}\text{Ra})T)] / \lambda(^{228}\text{Ra})}{A_0(^{226}\text{Ra}) [1 - \exp(-\lambda(^{226}\text{Ra})T)] / \lambda(^{226}\text{Ra})} \\ &= 3 \times \frac{[1 - \exp(-\lambda(^{228}\text{Ra})T)] / \lambda(^{228}\text{Ra})}{[1 - \exp(-\lambda(^{226}\text{Ra})T)] / \lambda(^{226}\text{Ra})} \end{aligned}$$

The amounts of ^{228}Ra and ^{226}Ra in the underground water supplied by the source rock will increase along their saturation curves, respectively, and will reach their saturation. Since the half-life of ^{228}Ra , 5.8 y, is smaller than that of ^{226}Ra , 1.6×10^3 y, the activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ will decrease with increasing residence time of the water within the aquifer until 7 000 y and will then level off at an equilibrium level with $^{232}\text{Th}/^{238}\text{U}$ activity ratio of the source rock as

shown in Fig. 3. As the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio may possibly vary with the residence time of the water in the aquifer, the activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ can provide the age of an underground water.

The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in hot-spring waters in Table 3 tended to be smaller than those in river waters. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in spring waters in Morocco was also smaller than those in river waters^{5), 22)}. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios for the 2 hot-spring waters given in Table 3 staying in deeper aquifer of 300 m and 1 100 – 1 500 m deep with possibly longer residence time may be smaller than those of waters in shallow aquifer, which may be the source of river waters with short residence time. If an age determination is al-

lowed to be made on the basis of this assumption, the ages of Nomekoi-no-yu and Tsuru-no-yu hot-spring waters may be estimated to be 30 and 35 years, respectively.

3.3 Variation in $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in river waters with precipitation

Figure 4 shows the variation in the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in Asakawa river from Feb. 2002 to Dec. 2003 and Nippara-gawa river from Jan. 2002 to Jan. 2003 along with the amount of daily precipitation. The correlation between the $^{228}\text{Ra}/^{226}\text{Ra}$ in river waters with the amount of precipitation was poor. Figure 5 shows the variation in $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in Asakawa river with the total amount of precipitation within 1 day and within 3 days pre-

Fig. 4 $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters and the amount of the precipitation in the period from 2002 to 2003.

■ : $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio, ○ : Amount of precipitation

Fig. 5 Variation in $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters with the amount of the precipitation.

□ : Amount of precipitation within 1 day
● : Amount of precipitation within 3 days

ceding to the observations of the activity ratios. When the total amount of precipitation was less than 20 mm, $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio decreased rapidly from 3 to 2 with an increase in

the amount of the precipitation, while the amount of the precipitation exceeded 20 mm, the $^{228}\text{Ra}/^{226}\text{Ra}$ maintained the value of 2.

The amount of precipitation at the present site is small in winter and large in summer. The observation was carried out in the periods from Dec. 2003 to Jan. 2004 for winter season and May to June 2004 for summer season. Activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ in the river waters was from 2.0 to 2.9 in winter and from 1.7 to 2.7 in summer, and the decreased activity ratios with rainfalls tended to recover afterward as shown in Figs. 6(a) and (b).

Kuramochi¹⁶⁾ reported that the flow rate of Tsuru-no-yu hot-spring waters increased with increasing amount of the precipitation and showed its maximum several hours after a heavy rainfall. Discharge of Ubako hot-spring water from Hakone caldera was also reported to increase with increase in the amount of the

Fig. 6 $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters and the amount of the precipitation in the period from 2003 to 2004.

— : $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters, ○ : Amount of precipitation

precipitation²³⁾. Oyama and Mizuno²⁴⁾ suggested that a rise of underground water level caused by a heavy rainfall enhanced the discharge of the hot-spring water.

As shown in Fig. 5, the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters tended to be small after the rainfall heavier than 20 mm/d. The reduced $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in river waters suggests that heavy rainfalls forced the deeper or older underground water with a longer residence time to discharge into river water to reduce the activity ratio.

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References

- 1) Kraemer, T. F. and Curwick, P. B., Radium isotopes in the lower Mississippi river, *J. Geophys. Res.*, **96**, 2797-2806 (1991)
- 2) Hakam, O. K., Choukri, A., Reyss, J.-L. and Lferde, M., Determination and comparison of uranium and radium isotopes activity ratios in samples from some natural water sources in Morocco, *J. Environ. Radioact.*, **57**, 175-189 (2001)
- 3) Eikenberg, J., Tricca, A., Vezzu, G., Bajo, S., Ruethi, M. and Surbeck, H., $^{228}\text{Ra}/^{226}\text{Ra}/^{224}\text{Ra}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ isotope relationship for determining interactions between ground water and river water in the upper Rhine valley, *J. Environ. Radioact.*, **54**, 133-162 (2001)
- 4) Tomoyose, N., Tanahara, A., Takemura, M., Toguchi, A. and Taira, H., Radioisotopic study on the ground waters of Okinawa Island, Japan, *Chikyukagaku (Geochemistry)*, **31**, 227-233 (1997) (in Japanese with English abstract)
- 5) Sturchio, N. C., Banner, J. L., Binz, C. M., Heraty, L. B. and Musgrove, M., Radium geochemistry of ground waters in Paleozoic carbonate aquifer, mid-continent, USA, *Appl. Geochem.*, **16**, 109-122 (2001)
- 6) Zhuo, W., Ikeda, T. and Yang, X., Occurrence of ^{222}Rn , ^{226}Ra , ^{228}Ra and U in groundwater in Fujian Province, China, *J. Environ. Radioact.*, **53**, 111-120 (2001)
- 7) Yamamoto, M., Toguchi, A., Sashimono, K., Komura, K., Ueno, K. and Sakanoue, M., ^{210}Pb and ^{210}Po in volcanic products and mineral springs (2)—Measurement of ^{210}Pb and ^{210}Po contents in mineral springs —, *J. Balneol. Soc. Jpn.*, **44**, 18-27 (1994) (in Japanese with English abstract)
- 8) Saito, T., Ueda, H., Ohta, T. and Sato, J., Determination of radium isotopes concentrations in hot-spring waters with cation exchange resin, *J. Balneol. Soc. Jpn.*, **52**, 3-11 (2002)
- 9) Kametani, K. and Matsumura, T., Determination of ^{238}U , ^{234}U , ^{226}Ra and ^{228}Ra in spring waters of Sanin district, *RADIOISOTOPES*, **32**, 18-21 (1983) (in Japanese with English abstract)
- 10) Yang, H., Hwang, D. and Kim, G., Factor controlling excess radium in the Nakdong river estuary, Korea: Submarine groundwater discharge versus desorption from riverine particles, *Mar. Chem.*, **78**, 1-8 (2002)
- 11) Krest, J. M., Moore, W. S. and Rama, ^{226}Ra and ^{228}Ra in the mixing zones of the Mississippi and Atchafalaya rivers: indicators of groundwater input, *Mar. Chem.*, **64**, 129-152 (1999)
- 12) Sakai, A., Geology of the Itsukaichi district. With geological sheet map at 1 : 50,000, *Geol. Surv. Jap.* (1987) (in Japanese with English abstract)
- 13) Mitsuchi, T., Explanatory text of the geological map of Japan at 1 : 75,000, Imperial geology survey of Japan (1932) (in Japanese with English abstract)
- 14) Omori, M. ed., *Nichiyo-no-chigaku*, Tsukiji shokan (1989) (in Japanese)
- 15) Kanroji, Y., Tanaka, S. and Ichiyanagi, H., Regional distribution and water qualities of hot-springs distributed from Tama to Okutama area in Tokyo Metropolis, *J. Balneol. Soc. Jpn.*, **27**, 32-46 (2000) (in Japanese with English abstract)
- 16) Kuramochi, F., On the Tsuruno-yu hot-spring, *J. Geography*, **65**, 46 (1956) (in Japanese with English abstract)
- 17) Takaoka, Y., Geological study of Okutama-cho, Nishitama-gun, Tokyo-to and the vicinity of Ten-

- shozan, *Tohogakuho*, **30**, 127-162 (1980) (in Japanese with English abstract)
- 18) Nagase, K., *Table of chemical constituents for Nomekoi-no-yu* (1997)
- 19) Hagiwara Boring Co. Ltd., *Priv. comm.* (2005)
- 20) Moore, W. S., Sampling ^{228}Ra in the deep ocean, *Deep-Sea Res.*, **23**, 647-651 (1976)
- 21) Ohta, T., Saito, T. and Sato, J., Collection of radium isotopes in natural waters by manganese-impregnated acrylic fiber, *RADIOISOTOPES*, **53**, 1-11 (2004)
- 22) Hakam, O. K., Choukri, A., Moutia, Z., Chouak, A., Cherkaoui, R., Reyss, J. -L. and Lferde, M., Uranium and radium in groundwater and surface water samples in Morocco, *Radiat. Phys. Chem.*, **61**, 635-654 (2001)
- 23) Imata, H., Oyama, M., Seino, T. and Yamaguchi, N., Variation of water level, temperature and discharge on hot springs in Hakone volcano, *Proc. 38th Conf. SITH*, Beppu, 270-274 (2004)
- 24) Oyama, M. and Mizuno, A., Numerical simulation of spring water discharge — Characteristics and applications of a nonlinear two-hole tank model —, *J. Groundwater Hydrology*, **36**, 1-11 (1994)

要 旨

東京都西部の奥多摩地域における河川水中の $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比

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東京都西部の奥多摩地域に流域を持つ河川水とその地域に湧出する温泉水及びこの地域を構成する岩石(堆積岩)の $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比の観測を行ったところ, 河川水中の $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比は $1.0 \sim 2.9$, 温泉水では $0.7 \sim 0.8$, 岩石では $1.2 \sim 1.7$ の範囲にあった。河川水中と岩石中の $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比の平均値はそれぞれ $2.0 \pm 0.0_3$ と 1.4 ± 0.1 であり, 河川水中の $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比は岩石中のものより高い傾向が見られた。地下深部にあって滞留時間が長いと考えられる温泉水中の $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比は河川水中のものより低い傾向にあった。河川水中の $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比は一日の降水量が 20 mm を超える強い降雨の後に減少する傾向がみられた。この $^{228}\text{Ra}/^{226}\text{Ra}$ 放射能比の減少は, 強い降雨により深部にある滞留時間の長い地下水が河川水中へ押し出された結果ではないかと考えられた。