

Concentrations of ^{226}Ra and ^{228}Ra in Hot Spring Waters from Tokyo Metropolis and Its Vicinity, Japan

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The average concentrations of ^{226}Ra and ^{228}Ra in the water in hot springs in and around metropolitan Tokyo were 39 mBq L⁻¹ and 55 mBq L⁻¹ for NaCl-type water and 0.6 mBq L⁻¹ and 0.8 mBq L⁻¹ for simple alkaline-type water, respectively. From the chemical characteristics, the hot spring waters from the Kazusa and Miocene formations are classified as NaCl and that from the Shimanto and Chichibu formations is simple alkaline. This indicates that the concentrations of Ra isotopes are high for the NaCl type and low for the simple alkaline type and are correlated with the relevant sedimentation layer from which the hot spring water is well up. Good correlation was observed for the ratios in the concentrations of Ra/Na and Ra/Cl in both types of hot spring water. The correlation patterns of Ra/Na and Ra/Cl for the simple alkaline type and sedimentary rock were different from those for the NaCl type and seawater, suggesting that the origin of the Ra isotopes may be different. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio was found to be related with Si concentration, showing that the activity ratio decreased with an increase of H₂SiO₃ for both the NaCl and the simple alkaline type. Assuming that the Ra isotopes in hot spring water were supplied from the relevant rock in the aquifer together with Si, the activity ratio of $^{228}\text{Ra}/^{226}\text{Ra}$ in hot spring water might be different between the NaCl and simple alkaline type, suggesting that the $(^{228}\text{Ra}/^{226}\text{Ra})_{t=0}$ activity ratio for residence time at $t = 0$ might be different for the two types.

1. Introduction

As ^{226}Ra and ^{228}Ra are progenies of ^{238}U and ^{232}Th , respectively, the concentrations of ^{226}Ra and ^{228}Ra in the water of hot springs is basically dependent on the U and Th content in the source rock of the aquifer. Since ^{230}Th and ^{232}Th , which are parents of ^{226}Ra and ^{228}Ra , are supposed to be insoluble in neutral alkaline-type hot spring water, ^{226}Ra and ^{228}Ra in the water are supplied from the relevant rock in the aquifer into the water by geochemical leaching and by α -recoil ejection from the Th isotopes. Assuming that the leaching efficiencies of ^{226}Ra and ^{228}Ra from the relevant rock into the hot spring water is equivalent, the variation of the concentrations of Ra isotopes in hot spring water might depend on the residence time of the hot spring water¹ or the mixing ratio of waters of different origins. Thus, the concentrations of ^{226}Ra and ^{228}Ra and the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio are considered useful tracers of the hydrological aspects of hot spring water.

However, the measurements of ^{226}Ra and ^{228}Ra in hot spring water were limited because of tiresome procedures of chemical separation from large amounts of sample water and difficulties in measuring low level Ra isotopes. Although by some workers determined wide ranges of ^{226}Ra and ^{228}Ra concentrations in hot spring water from Japan,²⁻⁴ the correlation of the concentrations of Ra isotopes and chemical components in hot spring water has not yet been well clarified. Nakano-Ohta and Sato⁵ classified hot spring water by correlations between the concentrations of ^{226}Ra and ^{228}Ra and the chemical constituents; the average concentrations of ^{226}Ra of some hot spring water from Japan reported in the literature were 104 mBq L⁻¹ for the NaCl type and 9 mBq L⁻¹ for the simple alkaline type. This indicated that the concentration of ^{226}Ra in the NaCl type was much greater than that in the simple alkaline type. The concentra-

tions of Ra isotopes in hot spring water from metropolitan Tokyo were determined with respect to the chemical constituents. The results indicated that the concentrations of Ra isotopes in the NaCl type averaged 70 times greater than those in the simple alkaline type,⁵ suggesting that the concentration of Ra isotopes depends highly on the chemical characteristics.

This paper deals with three investigations regarding Ra isotopes in the hot spring water of metropolitan Tokyo: (1) correlation of ^{226}Ra and ^{228}Ra in hot spring water with relevant rock in the aquifer from which the hot spring water is derived, (2) correlation of the concentration of Ra isotopes with major chemical components, and (3) the correlation of the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio with the concentration of H₂SiO₃.

2. Experiments

2.1. Sampling sites. Figure 1 shows the sampling sites of the hot springs and the geology of the studied areas.⁶ The hot springs in metropolitan Tokyo are grouped into three localities: (1) the core metropolitan area, (2) the Tama area, and (3) the Okutama area.⁷ The chemical characteristics of the water from the hot springs have been classified into (1) NaCl and NaHCO₃ types for the core metropolitan area, (2) NaCl and NaHCO₃ types for the Tama area, and (3) the simple alkaline type for the Okutama area. The evaporated residual materials of the hot spring water ranged from 98 to 580 mg L⁻¹ for the Okutama area,⁸ 150 to 2053 mg L⁻¹ for the Tama area,^{7,8} and 2,000 to 32,050 mg L⁻¹ for the core metropolitan area.⁸ This means that the chemical components in the hot spring water in the core metropolitan area are more abundant than those in the Okutama area.

2.2. Determination of Ra isotopes. Radium isotopes in the samples of hot spring water were quantitatively collected by Mn-impregnated acrylic fiber columns⁴ having a flow rate of less than 2 L min⁻¹. The column consisted of three small columns (3.5 cm ϕ , 90 cm long) of 31 g of Mn-impregnated acrylic

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fiber connected in series. This collection procedure required 2.5 hours for 100 L of water.

The Mn-impregnated acrylic fibers in the first and second columns were combined and packed into one airtight tin canister (76 mmφ, 24 mm H); the third column was packed into another canister. The canisters were stored for 1 month for γ -ray spectrometry so that ^{214}Pb and ^{228}Ac reached radioactive equilibrium with their parents, ^{226}Ra and ^{228}Ra , respectively. The radioactivity of ^{214}Pb and ^{228}Ac were determined by 351 keV and 911 keV γ -rays, respectively, using a Ge detector with 10% relative efficiency. Details of γ -ray spectrometry are

given by Saito et al.⁴

The concentrations of ^{226}Ra and ^{228}Ra were calculated by the amounts of ^{226}Ra and ^{228}Ra activities collected by the combined two Mn-impregnated acrylic fibers, and the sample volumes employed. Details of the procedure for determining ^{226}Ra and ^{228}Ra are given in the paper of Nakano-Ohta and Sato.⁵

2.3. Determination of major elements. Hot spring water from Oyedo-onnsen-monogatari, Seta-sanga-no-yu, Jindaiji-yukari, Kosuge-no-yu, and Hisamatsu were filtered with a membrane filter having a pore size of 0.45 μm , and the water was diluted with deionized water. The concentrations of Na,

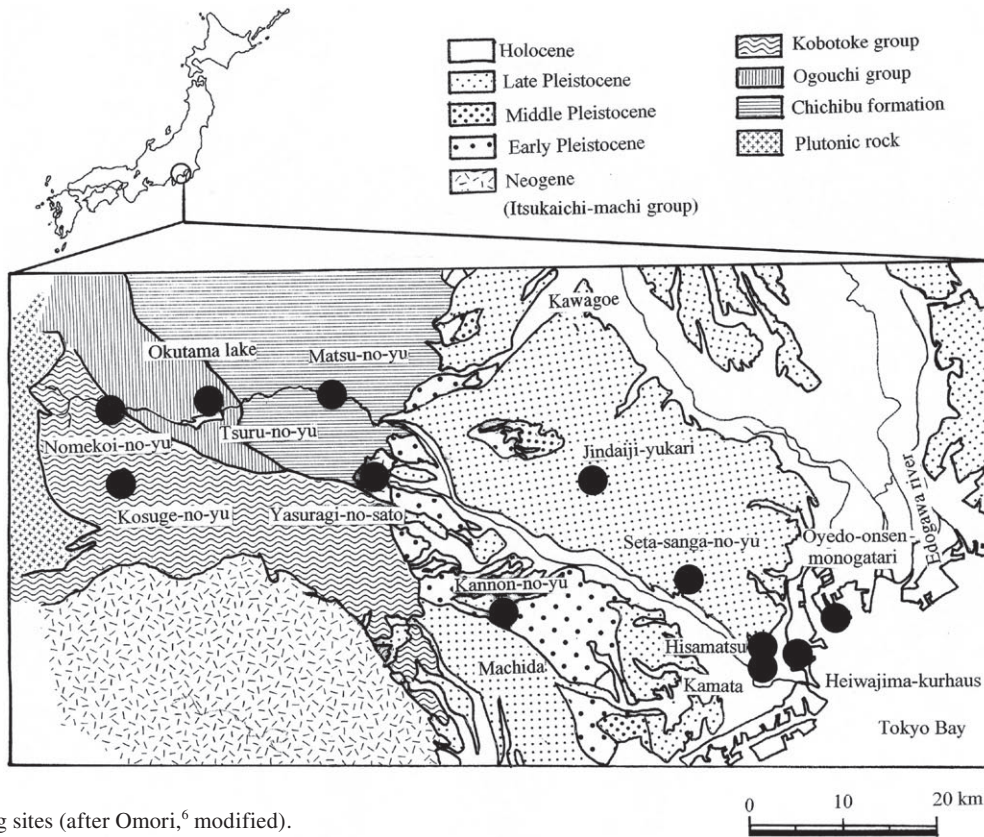


Figure 1. Sampling sites (after Omori,⁶ modified).

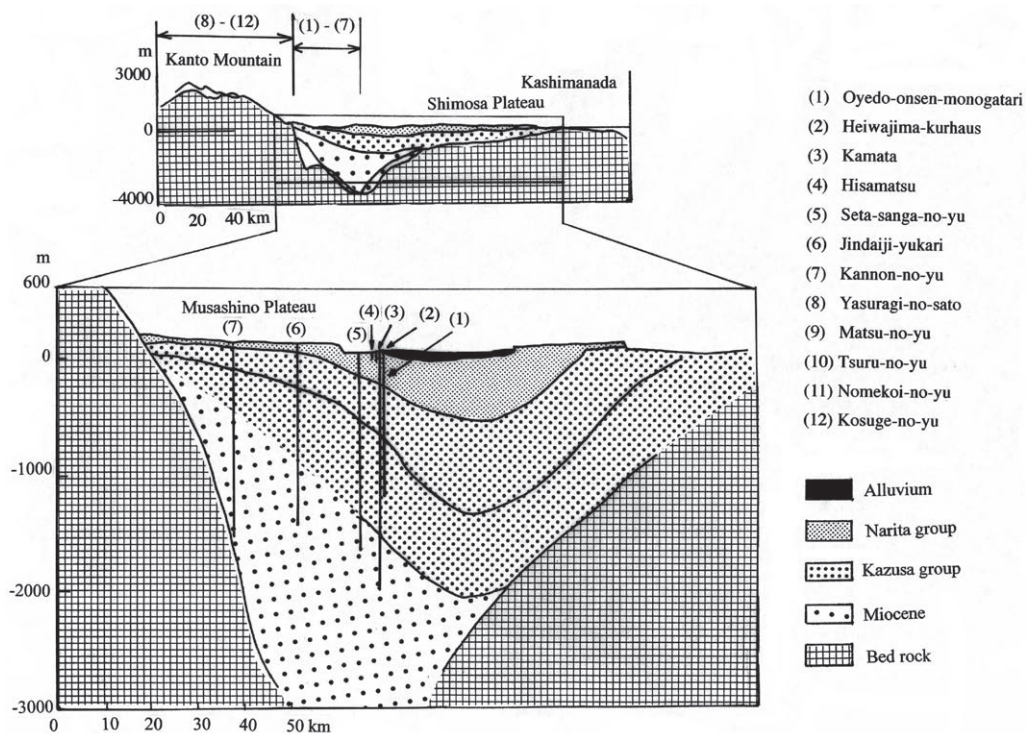


Figure 2. Geological map (after Kanroji⁸ and Kaizuka,^{9,10} modified) and the depth of bore-hole for some hot-spring waters.

K, Mg, and Ca in the hot spring water were determined by ICP-AES, ICPS7500 (Shimadzu Co. Ltd.). The analytical lines used for the ICP-AES measurements were 588.995 nm for Na, 766.491 nm for K, 279.553 nm for Mg, and 393.366 nm for Ca. Alternatively, the concentration of Cl was determined by ion chromatography, IC-7000 (Yokogawa Co. Ltd.) with an Excelpak ICS-A23 anion-exchange resin column. The analytical conditions for Cl were 3.0 mM of Na_2CO_3 for the eluent and 15 mM of H_2SO_4 for the regenerating solution. The elution speed of the sample water was 1.0 mL min^{-1} .

The concentrations of the major elements of hot spring water from Heiwajima-kurhaus, Kamata, Kannon-no-yu, Nomekoi-no-yu, Matsu-no-yu, and Tsuru-no-yu and the Si concentration were referred to the notice board of the chemical constituents for each hot spring. The chemical components were assumed constant from the day of determination of the major chemical components to our sampling day.

3. Results and Discussion

3.1. Correlation of Ra concentration and sedimentation layer of the aquifer. Table 1 lists the concentrations of ^{226}Ra and ^{228}Ra in the hot spring waters classified by chemical constituent and depth of borehole. The concentrations of Ra isotopes in the hot spring waters ranged from 0.3 to 84 mBq L^{-1} for ^{226}Ra and from 0.2 to 104 mBq L^{-1} for ^{228}Ra , respectively. The average concentrations were 39 mBq L^{-1} for ^{226}Ra and 55 mBq L^{-1} for ^{228}Ra in the NaCl type ($n = 5$) and 0.6 mBq L^{-1} for ^{226}Ra and 0.8 mBq L^{-1} for ^{228}Ra in the simple alkaline type ($n = 5$). This indicated that the Ra isotope concentration in the NaCl-type hot spring was 70 times greater than that in the simple alkaline type.

Hot springs are generally classified into two groups: (1) non-volcanic hot springs (fossil seawater or deep underground water) and (2) volcanic hot springs. The hot springs in the core metropolitan and Tama areas were reported to be non-volcanic hot springs; those in the eastern part of Yamanashi prefecture near the Okutama area were either non-volcanic or volcanic hot springs.⁸ Figure 2 shows the geology and the depth of the bore-

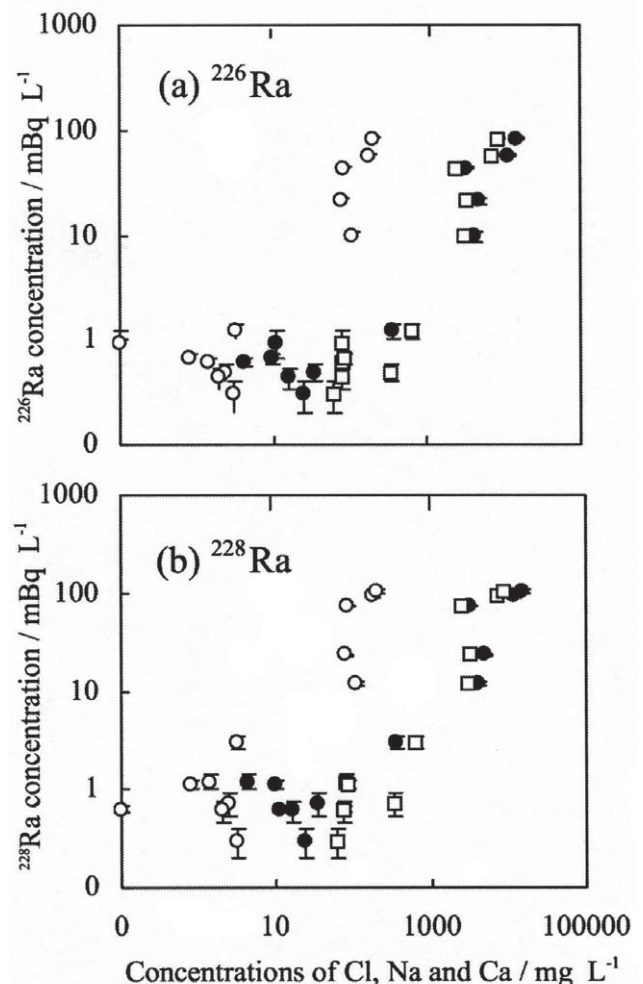


Figure 3. (a) Correlations between ^{226}Ra and Cl, Na, and Ca in hot spring waters. (b) Correlations between ^{228}Ra and Cl, Na, and Ca in hot spring waters. \circ : Ca-Ra, \bullet : Cl-Ra, and \square : Na-Ra.

TABLE 1: Concentrations of ^{226}Ra and ^{228}Ra , and $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in hot spring waters from Tokyo area

Chemical character	Sampling date	$^{226}\text{Ra} / \text{mBq L}^{-1*}$	$^{228}\text{Ra} / \text{mBq L}^{-1*}$	$^{228}\text{Ra} / ^{226}\text{Ra}$	Depth**/ m
NaCl type					
Oyedo-onsen-monogatari ^a	Dec. 9, 2005	84 ± 2	104 ± 4	1.2 ± 0.1	1300
Heiwajima-kurhaus ^a	Dec. 26, 2004	58 ± 2	94 ± 4	1.9 ± 0.1	2000
Kannon-no-yu ^b	Apr. 3, 2005	43 ± 1	73 ± 1	1.7 ± 0.1	1500
Seta-sanga-no-yu ^a	Dec. 6, 2005	21 ± 0.5	23 ± 1	1.1 ± 0.1	1700
Jindaiji-yukari ^a	Jul. 4, 2005	10 ± 0.2	12 ± 1	1.2 ± 0.1	1500
Tennyo-no-yu ^d	Aug. 8, 2003	18 ± 1	21 ± 1	1.3 ± 0.1	1600
Average		39	55	1.4	
NaHCO₃ type					
Kamata ^a	Dec. 26, 2004	1.2 ± 0.2	3.0 ± 0.4	2.5 ± 0.5	100
Hisamatsu ^a	Jul. 2, 2005	0.49 ± 0.09	0.73 ± 0.20	1.5 ± 0.5	60
Average		0.85	1.9	2.0	
Simple alkaline type					
Nomekoi-no-yu ^c	May 14, 2003	0.30 ± 0.08	0.24 ± 0.04	0.8 ± 0.3	1300–1500
Tsuru-no-yu ^c	May 20, 2003	0.94 ± 0.26	0.63 ± 0.04	0.7 ± 0.2	300
Matsu-no-yu ^c	May 14, 2003	0.66 ± 0.08	1.1 ± 0.11	1.7 ± 0.3	30
Kosuge-no-yu ^c	Jul. 12, 2005	0.43 ± 0.10	0.62 ± 0.15	1.4 ± 0.5	1500
Yasuragi-no-sato ^c	May 10, 2005	0.62 ± 0.06	1.2 ± 0.2	1.9 ± 0.2	1300–1500
Average		0.6	0.8	1.3	

*Errors represent the counting statistics.

**The depth of bore-hole of hot-spring.

^aMetropolitan area. ^bTama area. ^cOkutama area. ^dYamanashi Pref.

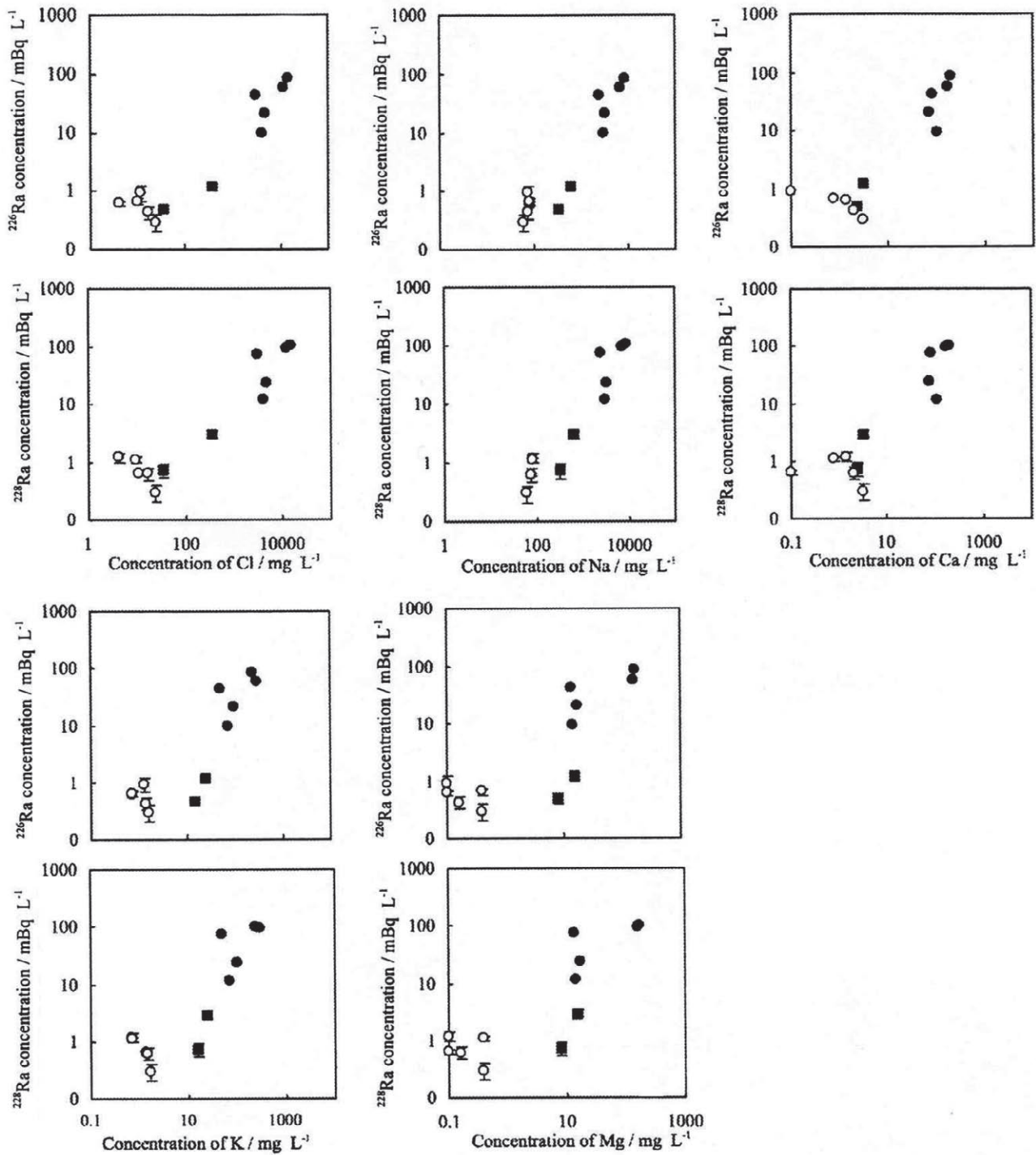


Figure 4. Correlation between ^{226}Ra and ^{228}Ra , and Cl, Na, Ca, K, and Mg in hot spring waters. ●: NaCl type, ■: NaHCO_3 type, and ○: Simple alkaline type.

holes of the hot springs at Kanroji⁸ and Kaizuka.^{9,10} The Kazusa and Miocene formations are relevant sedimentation layers for Oyedo-onsen-monogatari, Heiwajima-Kurhaus, Seta-sanga-no-yu, Jindaiji-yukari, and Kannon-no-yu whereas the Shimanto and Chichibu formations are relevant for Nomekoi-no-yu, Tsuru-no-yu, Matsu-no-yu, Kosuge-no-yu, and Yasuragi-no-yu.⁷⁻¹⁴ The chemical characteristics of the Kazusa and Miocene formations are the NaCl type whereas the Shimanto and Chichibu formations are the simple alkaline type, indicating that different concentrations of Ra isotopes exist in the different aquifers from which the hot spring water is welled up. Seki et al.¹⁵ reported that the chemical characteristics of well water from depths of 700–2000 m in the eastern Kanto plain of Japan were classified according to the surrounding geology as follows. (1) The hot spring waters from the Kanto plain were of the NaCl type, and consisted of fossil seawater. (2) Those from the marginal mountainous area were of

the alkaline type with a low concentration of NaHCO_3 , which might consist of meteoric water reacting with silicic igneous rocks. (3) Moderate alkaline concentration Na/Ca- SO_4 type mostly from the Kinugawa graben, which might consist mainly of meteoric water reacting with pre-Tertiary sedimentary rocks and Tertiary volcaniclastic rocks. The hot springs studied in the present work and observed by Seki et al.¹⁵ were collected from the coastal area of the Pacific Ocean to the mountainous area of the central part of the Kanto plain. The area we studied is located in the western part of the Kanto plain, while the eastern part of the area was observed by Seki et al.¹⁵ The chemical characteristics of the presently observed hot springs were similar to those that appeared in the literature:¹⁵ a high concentration of chemical components with the NaCl type from the Kanto plain area and a low concentration of components with the alkaline type from the Kanto mountainous area. If the concentration of Ra isotopes in hot spring water depends on the

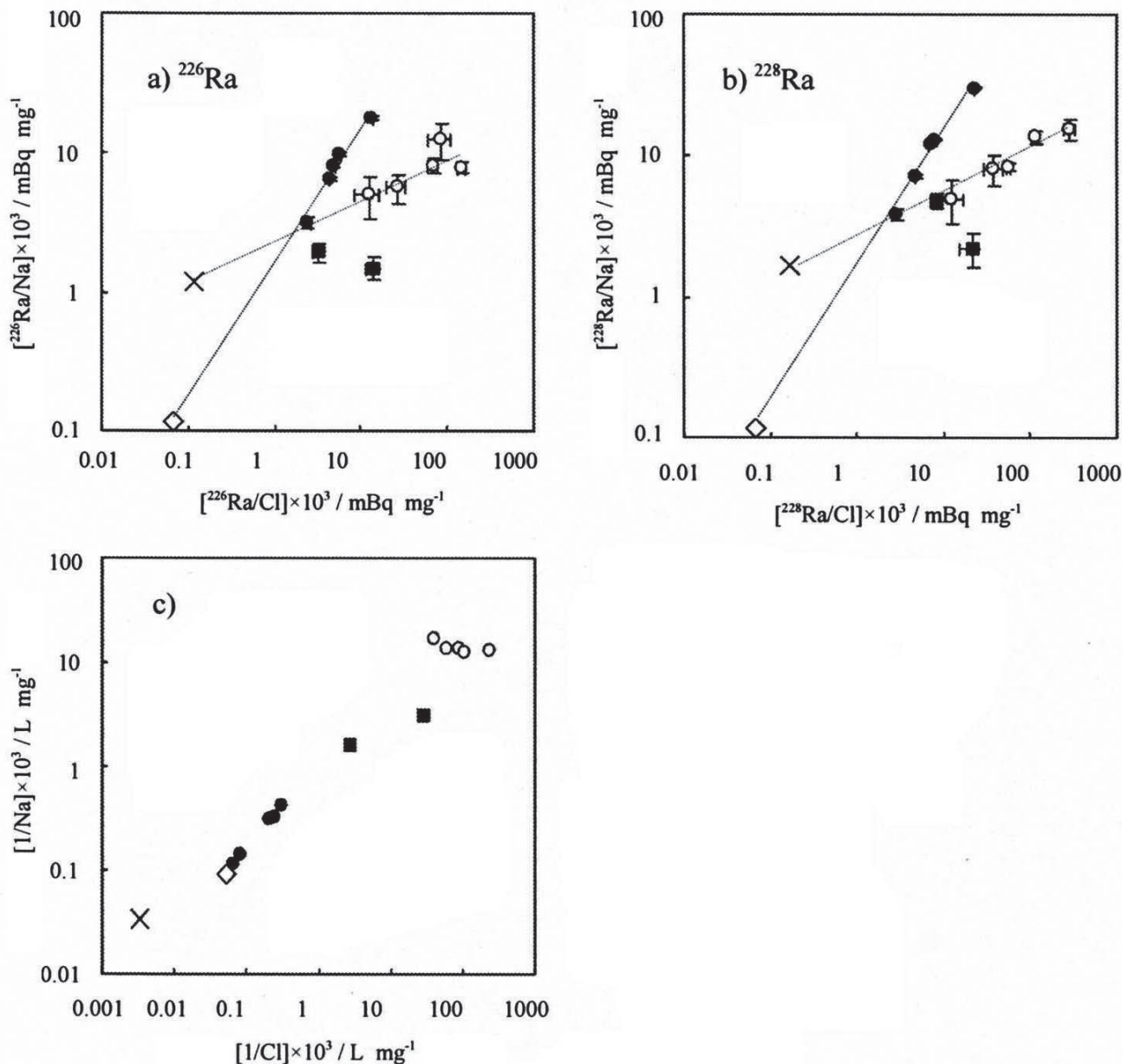


Figure 5. (a) Relation between $^{226}\text{Ra}/\text{Na}$ and $^{226}\text{Ra}/\text{Cl}$ in hot spring waters. (b) Relation between $^{228}\text{Ra}/\text{Na}$ and $^{228}\text{Ra}/\text{Cl}$ in hot spring waters. (c) Relation between $1/\text{Na}$ and $1/\text{Cl}$ in hot spring waters. ●: NaCl type, ■: NaHCO_3 type, ○: Simple alkaline type, ◇: seawater, and ×: sedimentary rock.

sedimentation layer, the hot spring water from the Kazusa group and the Narita group in the Kanto plain may contain a high level of Ra isotopes, as observed in the present study. The concentrations of Ra isotopes in three areas of underground water in Mobara, Chiba prefecture, in the eastern part of the Kanto plain along the coast of the Pacific Ocean were measured to be as high as those in the core metropolitan area. Details concerning the Mobara observation will be given elsewhere.

3.2. Correlation of Ra concentration and chemical constituents. The concentrations of ^{226}Ra and ^{228}Ra in hot spring waters increased with an increase in the concentrations of Cl, Na, and Ca, as shown in Figure 3. Figure 4 shows the correlations between the observed concentrations of ^{226}Ra and ^{228}Ra , and among Na, K, Ca, Mg, and Cl. The concentrations of the Ra isotopes for the NaCl type increased with an increase in the concentrations of K, Mg, Na, Ca, and Cl, while those for the simple alkaline type increased with an increase in the Na concentration, but decreased with an increase in the concentrations of K, Mg, Ca, and Cl.

Figures 5(a) and 5(b) show the correlations of the ratios of $[\text{}^{226}\text{Ra}/\text{Na}]$ and $[\text{}^{226}\text{Ra}/\text{Cl}]$, and $[\text{}^{228}\text{Ra}/\text{Na}]$ and $[\text{}^{228}\text{Ra}/\text{Cl}]$ for the NaCl type, simple alkaline type, NaHCO_3 type, seawater, and

sedimentary rock, respectively. Figure 5(c) shows the correlation of the ratios of $[1/\text{Na}]$ with $[1/\text{Cl}]$. A good correlation between $[1/\text{Na}]$ and $[1/\text{Cl}]$ was observed for the NaCl-type hot spring. The concentrations of ^{226}Ra and ^{228}Ra in surface seawater from Tokyo Bay in the Pacific Ocean ranged from 1.1 to 1.9 mBq L^{-1} for ^{226}Ra and from 0.9 to 1.4 mBq L^{-1} for ^{228}Ra , respectively.¹⁶ Although the concentrations of ^{226}Ra and ^{228}Ra in the surface seawater in the Pacific Ocean are higher than those just offshore,¹⁷ those in the surface seawater of Tokyo Bay are on the order of 1 mBq L^{-1} .^{16,17} The ratios of concentration of surface seawater were 1×10^{-4} and 0.7×10^{-4} for $[\text{}^{226}\text{Ra}/\text{Na}]$ and $[\text{}^{226}\text{Ra}/\text{Cl}]$, and 1×10^{-4} and 0.7×10^{-4} for $[\text{}^{228}\text{Ra}/\text{Na}]$ and $[\text{}^{228}\text{Ra}/\text{Cl}]$, respectively. The ratios of concentration of $[\text{Ra}/\text{Na}]$ and $[\text{Ra}/\text{Cl}]$ for seawater are also plotted in Figures 5(a) and 5(b) with an open square. The average concentrations of ^{226}Ra and ^{228}Ra in sedimentary rock,¹⁸ which may be the relevant rock in the aquifer for Nomekoi-no-yu, Matsu-no-yu, Tsuru-no-yu, and Kosuge-no-yu, were 36 mBq g^{-1} and 50 mBq g^{-1} , respectively. The ratios of concentration of sedimentary rock were 1×10^{-3} for $[\text{}^{226}\text{Ra}/\text{Na}]$, 1×10^{-4} for $[\text{}^{226}\text{Ra}/\text{Cl}]$, 2×10^{-3} for $[\text{}^{228}\text{Ra}/\text{Na}]$, and 2×10^{-4} for $[\text{}^{228}\text{Ra}/\text{Cl}]$ as shown in Figure 5(a).

The ratios of concentration of $[\text{}^{226}\text{Ra}/\text{Na}]$ and $[\text{}^{226}\text{Ra}/\text{Cl}]$ and

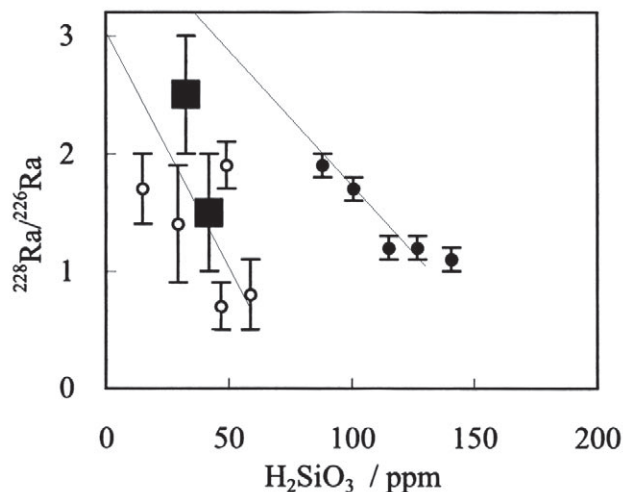


Figure 6. Correlation between $^{228}\text{Ra}/^{226}\text{Ra}$ and H_2SiO_3 in hot-spring waters. ●: NaCl type, ■: NaHCO_3 type, and ○: Simple alkaline type.

of $[^{228}\text{Ra}/\text{Na}]$ and $[^{228}\text{Ra}/\text{Cl}]$ showed good correlation both for the NaCl type and simple alkaline type, as shown in Figure 5(a). The extrapolated values of the ratios of $[\text{Ra}/\text{Na}]$ to $[\text{Ra}/\text{Cl}]$ for the NaCl type agreed with those for seawater, while the simple alkaline type agreed with the ratio of $[\text{Ra}/\text{Na}]$ and $[\text{Ra}/\text{Cl}]$ of the sedimentary rock. The Ra isotopes in the NaCl-type hot spring water might be supplied from 'fossil seawater' in the sedimentation layer, while those in the simple alkaline-type hot spring water might be supplied from the sedimentary rock by meteoric water.

3.3. $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in the water of hot springs.

The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio ranged from 0.7 to 2.5, as shown in Table 1. Although the concentrations of ^{228}Ra and ^{226}Ra had a wide range, the hot springs could be classified by chemical characteristics, however, the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio but not by the concentrations of the major elements. However, the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio was found to be closely related with the concentration of H_2SiO_3 , as shown in Figure 6. Namely, the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio decreases with an increase of the H_2SiO_3 concentration for both the NaCl type and simple alkaline type of hot spring. As H_2SiO_3 in hot spring water is considered to be leached from the rock in the aquifer, the activity ratios may be classified by H_2SiO_3 concentration in both the NaCl and simple alkaline types, as shown in Figure 6. Assuming that the Ra isotopes in hot spring water were supplied from the rock in the aquifer together with Si, the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio may decrease with an increase of the residence time of the hot spring water in the rock in the aquifer. An $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio for a 0 ppm concentration of Si indicates the initial $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio at time zero. Based on the assumption stated above, the initial $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of hot spring water for the NaCl type may be different from the simple alkaline type, that is, the activity ratio, $(^{228}\text{Ra}/^{226}\text{Ra})_{t=0}$, of the NaCl-type hot spring is higher than that of the simple alkaline type.

4. Conclusion

The concentrations of ^{226}Ra and ^{228}Ra in the hot spring water from metropolitan Tokyo suggested that:

(1) The chemical characteristics of the hot spring waters from the Kazusa and Miocene formations is the NaCl type and that from the Shimanto and Chichibu formations is the simple alkaline type, from the fact that the concentrations of Ra isotopes are high for the NaCl type and low for the simple alkaline type, and correlate well with the relevant sedimentation layer from which the hot spring water is derived.

(2) Although the concentration ratios of $[\text{Ra}/\text{Na}]$ and $[\text{Ra}/\text{Cl}]$ in hot spring waters showed good correlation, those for the simple alkaline type and sedimentary rock were different from those for the NaCl type and seawater, suggesting that the origin of the Ra isotopes for the simple alkaline type is different from that for the NaCl type.

(3) The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio decreased with increasing H_2SiO_3 concentration for both the NaCl and simple alkaline types. Assuming that the Ra isotopes were leached from the relevant sedimentary rock together with Si, $(^{228}\text{Ra}/^{226}\text{Ra})_{t=0}$ for the simple alkaline-type hot spring water might be different from that of the NaCl-type hot spring water.

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