Radioactive Disequilibrium of Uranium and Thorium Nuclide Series in Hot Spring and River Water from Peitou Hot Spring Basin in Taipei

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The distributions of the naturally occurring radionuclides ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{228}Ra, and ^{226}Ra in river and hot spring water within the Peitou hot spring basin at the northern part of Taiwan were studied by using alpha- and gamma-spectrometric methods and liquid scintillation counting method. The respective concentration ranges of ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th, and ^{226}Ra in river and hot spring water were 0.80–48, 1.2–51, 0.05–44, 0.09–39, 0.19–342, 3.0–22.5 mBq/L and 2.3–85, 2.9–91, 0.67–73, 1.14–66, 1.01–630, and 4.5–36.5 mBq/L, respectively. In general, the distributions of uranium-series and thorium-series nuclides in river water, hot spring water were all pH dependent. The respective radioactivity ratios of ^{234}U/^{238}U in river and hot spring water are observed to be 1.0–1.5 and 1.03–1.5, however, the radioactivity ratios of ^{228}Th/^{232}Th in river and hot spring water are 1.4–19 and 1.5–8.8, respectively. The high ^{228}Th/^{232}Th activity ratios showed that besides the recoil effect, enrichment of ^{228}Ra to ^{232}Th in the waters was another possible factor causing an excess of ^{228}Th. Because radium may exist as (Pb, Ba, Ra)$(SO_4)_X$ pseudo-colloid in hot spring water, which is the same as the more famous mineral of Peitou-stone (Hukutolite), therefore, ^{228}Ra and ^{226}Ra are more abundant in the hot spring sediments, which radioactivity are18–3010 and 16–230 Bq/kg (dry), respectively.

Introduction

The Peitou hot spring basin is located in the northern part of Taiwan (see Figure 1). There are volcanic vestiges and hot springs all around this area. Hot springs here spread over the western part of Taiwan between two geological faults. There are three main streams, Waishuang, Nanhuang and Huangkang Streams, running through this area, emptying into the Keelung River, and finally emptying into the Taiwan Strait through the Tamsui River. Owing to a violent material exchange between the subterranean hot waters and the rock stratum, the erosion of the rocks is greatly increased by the high-acidic sulfurous waters.

After Okamoto, who first found the radioactive mineral, Hokutolite, in the Peitou hot spring in this area in 1907,[1] studies of the naturally occurring radionuclides in hot spring water of this area were undertaken one after another.[2–5] When a hot spring water abundant with naturally occurring radionuclides at high temperatures and its acidity empties into the rivers nearby, the distribution of the radionuclides may differ from river basin to river basin, since changes of the environment can cause fractionation between the naturally occurring radionuclides.

The purposes of this paper is to systematically study the distribution and migration of the naturally occurring radionuclides, ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{228}Ra and ^{226}Ra in the main rivers and hot springs of the Peitou hot spring basin by α-spectrometry, γ-ray spectrometry and liquid scintillation counting method.

Experimental

Sample Description. Figure 1 shows the sampling spots in the Peitou hot spring basin. All the water samples collected were placed in 20-L bottles containing 50 mL of HCl to avoid hydrolysis and radiocolloid formation. In addition, we collected 100mL of water without HCl, and cooled to room temperature to measure its pH value with a pH meter. All the temperatures recorded were made by a Hg-thermometer at the sampling spots. In the Waishuang Stream, we collected samples at six spots (V1 – V6). In the Nanhuang Stream, samples were collected at eight spots (N1 – N8). In the Huangkang Stream, at four spots (H1 – H4) were the samples collected. In addition, at the Peitou hot spring, we collected water samples at four spots (P1–P4) and sediment samples at two spots (P3 – P4). At the Hsingi-lu hot spring, we collected water samples at two spots (H1 – H2) and sediment samples at one spots (H2). At the Liuhuang-guu hot spring, one water sample (L1) and one sediment sample (L1) were collected. And, only one hot spring water sample (M1) was collected at the Matsao hot spring.

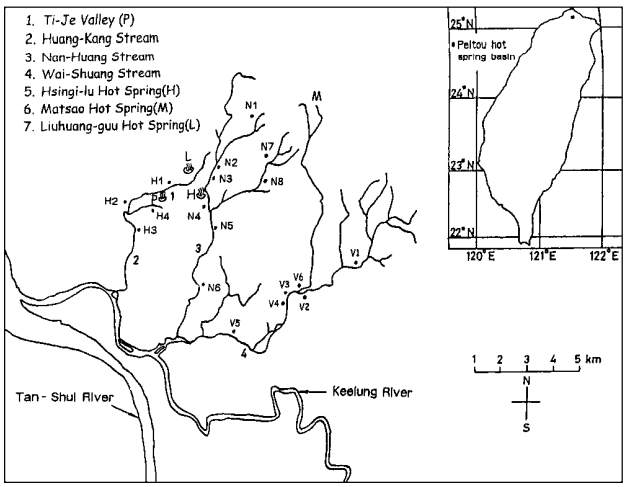

Figure 1. Sampling sites (•) in the Peitou hot spring basin in Taiwan. H: Huangkan stream, N: Nanhuang stream, V: Waishuang stream, P: Peitou hot spring, H: Hsingi-lu hot spring, L: Liuhuang-guu hot spring, M: Matsao hot spring.

Separation of Uranium.[6] ^{232}U was added as a tracer to each sample, followed by addition of CyDTA to mask the thorium in the sample. Uranium was sorbed on a Chelex-100 resin (Na-form; 50–100 mesh), and then desorbed by ammonium carbonate solution which was subjected to successive treatment with nitric, perchloric and sulfuric acids. The resulting

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solution was electrolyzed for 2 hours with platinum as an anode and a stainless steel plate as a cathode. The cathode was dried by an infrared lamp and cooled to room temperature for alpha counting.

Separation of Thorium.[7] The thorium was concentrated by coprecipitation of ferric hydroxide and then purified by an anion exchange resin column. ^{229}Th tracer was added to each sample, and thorium was collected as a ferric hydroxide precipitate. The precipitate was dissolved by nitric acid and then passed down a Dowex-1 × 8 resin column (NO_3^- form; 50–100 mesh) to remove thorium. Thorium was desorbed by hydrochloric acid, and treated by nitric, perchloric and sulfuric acids as in the case of uranium. The final solution was electrolyzed for 1.5 hours, and the stainless steel cathode was dried by an infrared lamp. After cooling, alpha activity was counted.

Alpha Counting. Alpha counting was done using a NUMELEC Model NU114 grid ionization chamber, and the counting efficiency of which was determined by means of an ^{241}Am standard source. From the chemical yield and the counting efficiencies of the instrument, the disintegration rates of the uranium and thorium isotopes were determined.

Separation and Determination of ^{226}Ra.[8] ^{226}Ra was concentrated and separated using the barium sulfate coprecipitation method and then the radioactivity of ^{226}Ra was determined by liquid scintillation counting.

Radium was coprecipitated with barium carrier as sulfate. After purifying the radium by recrystallization, the precipitate was dissolved in an EDTA solution. The purified ^{226}Ra in EDTA solution was packed into a 20-mL glass vial and then diluted to 10 mL with H_2O. After the addition of 10 mL of a POP–POPOP cocktail in toluence, the vial containing the sample was composed of two phases, was kept air-tight and allowed to stand for over 30 days to establish a radioactive equilibrium between ^{226}Ra and ^{222}Rn. The activity of ^{222}Rn was measured on a Packard Model 2560 TR/XR liquid scintillation counter within an energy window of 335–530 keV, from which the activity of ^{226}Ra was then determined. A 3.824-day half-life of ^{222}Rn was determined by an activity ingrowth-curve at 335–530 keV using a spiked ^{226}Ra water sample which was operated in the process mentioned above. Besides, a 100 % counting efficiency, 96.7 ± 2.3 % chemical yields of radium were estimated using ^{226}Ra spiked water samples which were operated in the same process.

Determination of ^{228}Ra. The hot spring sediment samples were heated to dry under 110 ˚C for 24 hours and kept air-tight in the acrylic containers for 30 days to establish radioequilibrium between ^{228}Ra and ^{228}Ac. The γ-ray activity of ^{228}Ac was counted using a pure Ge detector, and from which the radioactivity of ^{228}Ra was solved.

Results and Discussion

I. River Water. The analytical results of the radioactivities of the naturally occurring radionuclides of the three main rivers in the Peitou hot spring basin are shown in Table 1. Depending upon the mixing ratio of the hot spring water, the temperature and pH of the river water vary greatly; their ranges are 13˚C–47˚C and 1.80–7.64, respectively.

Radioactivity Distribution. The radioactivity distribution from some of the river water in certain areas varies greatly because the hot spring water is emptying into rivers in those areas.

1. Uranium. From Table 1, the concentration of uranium in the river water in the Peitou hot spring basin ranges from 0.064 to 3.8 ppb. The radioactivities of ^{238}U and ^{234}U were 0.80–48 and 1.2–51 mBq/L, respectively.

2. Thorium. The concentration of thorium in the river water of the Peitou hot spring basin ranges from 0.011 to 10.9 ppb, and the radioactivities of ^{232}Th, ^{230}Th, and ^{228}Th were 0.05–44, 0.09–39, and 0.19–342 mBq/L, respectively.

3. Radium. The river water investigated in this study, shows the concentration of ^{226}Ra ranges between 3.0 and 22.5 mBq/L.

It is likely the high uranium, thorium and radium contents of some river water samples collected in this area result from the contribution of the hot spring water.

pH Dependence. The chemical properties of uranium and thorium in the waters are mostly affected by the hydroxide. Therefore, the content of uranium and thorium in the river water is related to pH. Figure 2 shows the effects of the pH on the solubility of $UO_2(OH)_2$ and $Th(OH)_4$. The solubility shown in Figure 2 is based on 25 ˚C, μ = 0 and calculated from the solubility product (K_{sp}) of $UO_2(OH)_2$ and $Th(OH)_4$.[9] It differs from that of real river water, so Figure 2 is included just for reference. In Figure 2, obviously, $Th(OH)_4$ and $UO_2(OH)_2$ are soluble below pH = 5 and 7, respectively.

Figure 3 illustrates the concentration (ppb) of uranium and thorium in river water as a function of pH. In Figure 3, obviously, when pH ~7, the concentration of uranium and thorium is extremely low; when pH < 5, the concentration gradually increases. This indicates that, when pH ~7, uranium and thorium cannot be leached much from the fractures or pores of the rock in the form of hydroxide. When pH < 5, uranium and thorium are able to enter the river water in the form of UO_2^{2+} and Th^{4+} which are then suitable for leaching.

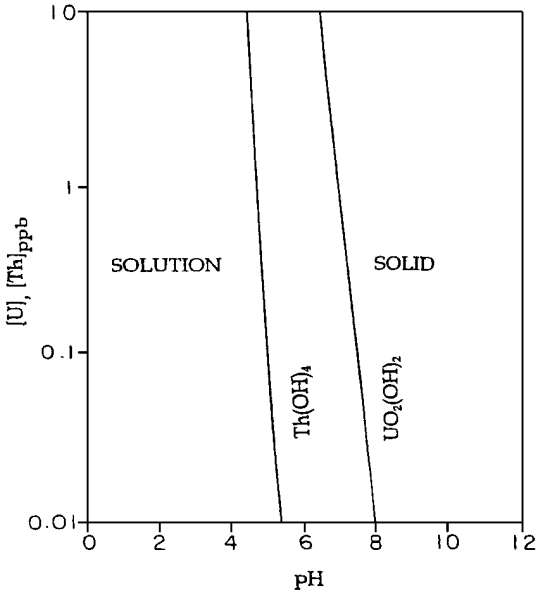

Figure 2. A plot of concentration of uranium and thorium vs. pH at 25 °C, μ = 0.

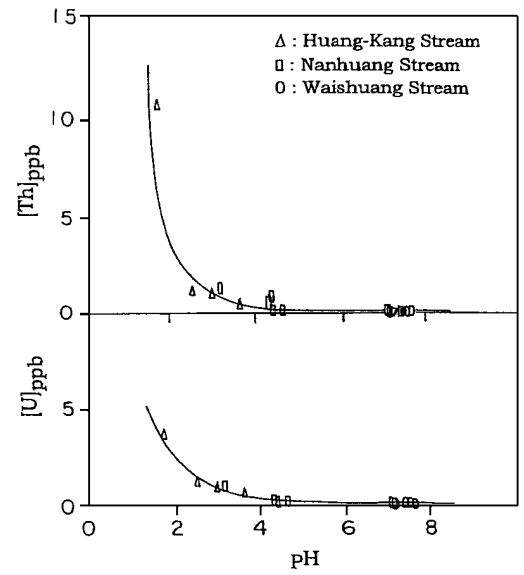

Figure 3. A plot of concentrations of U and Th vs. pH of river water.

TABLE 1: The Radioactivities of the Uranium and Thorium Family Members in the Stream Water

	Waishuang Stream						Nanhuang Stream								Huangkang Stream			
	Upstream		Downstream			Tributary	Upstream				Downstream		Tributary		Upstream		Downstream	Tributary
Location code	V1	V2	V3	V4	V5	V6	N1	N2	N3	N4	N5	N6	N7	N8	H1	H2	H3	H4
Temp. °C	13.0	13.0	14.0	13.0	14.0	13.5	15.5	18.0	17.5	22.0	21.0	21.0	42.0	28.0	21.0	19.5	23.0	47.2
pH	7.52	7.49	7.64	7.36	7.18	7.43	7.37	4.61	4.39	7.05	4.29	4.37	7.11	3.20	3.00	3.65	2.56	1.80
^{238}U (mBq/L)	1.6±0.1	1.5±0.1	1.4±0.1	1.4±0.1	1.4±0.1	0.80±0.09	2.0±0.2	2.5±0.2	2.0±0.2	1.9±0.2	3.5±0.3	2.6±0.3	1.4±0.1	12.6±0.8	10.6±0.6	7.5±0.5	14.2±0.7	48±2
^{234}U (mBq/L)	2.1±0.2	2.0±0.2	1.8±0.1	1.9±0.2	1.8±0.2	1.2±0.1	2.7±0.2	2.6±0.2	2.1±0.2	1.8±0.2	4.0±0.3	3.1±0.3	1.7±0.1	14.9±0.9	11.1±0.6	8.2±0.5	14.8±0.8	51±2
^{232}Th (mBq/L)	0.14±0.02	0.19±0.03	0.18±0.05	0.16±0.03	0.14±0.02	0.27±0.06	0.12±0.02	0.57±0.05	0.54±0.07	0.22±0.02	2.37±0.06	3.4±0.3	0.05±0.01	5.4±0.3	4.0±0.3	1.5±0.1	4.3±0.3	44±1
^{230}Th (mBq/L)	0.24±0.03	0.36±0.05	0.29±0.06	0.24±0.03	0.21±0.03	0.46±0.08	0.24±0.03	0.71±0.06	1.1±0.1	0.32±0.02	2.42±0.06	5.3±0.3	0.09±0.02	8.0±0.3	4.6±0.4	1.5±0.5	3.3±0.2	39±1
^{228}Th (mBq/L)	0.19±0.03	0.41±0.05	0.35±0.08	0.26±0.03	0.24±0.03	0.46±0.08	0.53±0.04	0.82±0.08	1.2±0.1	0.65±0.3	45.0±0.5	7.7±0.4	0.22±0.03	57±2	12.1±0.7	11.8±0.4	37±1	342±5
^{226}Ra (mBq/L)	4.6±0.1	6.4±0.1	7.6±0.2	6.1±0.2	7.9±0.2	5.11±0.07	5.9±0.1	4.1±0.1	7.1±0.2	5.4±0.2	14.8±0.4	3.0±1	7.5±0.3	16.8±0.5	5.5±0.1	6.00±0.06	11.9±0.3	22.5±0.7
U (ppb)	0.13±0.1	0.12±0.01	0.11±0.01	0.11±0.01	0.11±0.01	0.064±0.007	0.16±0.01	0.20±0.02	0.16±0.02	0.15±0.01	0.28±0.03	0. 21±0.02	0.11±0.01	1.01±0.06	0.86±0.05	0.60±0.04	1.14±0.06	3.8±0.2
Th (ppb)	0.035±0.006	0.047±0.008	0.05±0.01	0.039±0.006	0.035±0.005	0.07±0.02	0.029±0.005	0.14±0.01	0.13±0.02	0.053±0.04	0.59±0.02	0.85±0.07	0.011±0.003	1.34±0.07	0.99±0.08	0.37±0.03	1.07±0.07	10.9±0.3
U/Th (ppb/ppb)	3.71±0.7	2.±0.5	2.2±0.5	2.8±0.5	3.1±0.5	0.9±0.3	6±1	1.4±0.2	1.2±0.2	2.8±0.3	0.47±0.05	0.25±0.03	10±3	0.75±0.06	0.87±0.09	1.6±0.2	1.07±0.09	0.35±0.02
^{234}U/^{238}U	1.31±0.2	1.3±0.2	1.3±0.1	1.4±0.2	1.3±0.2	1.5±0.2	1. 4±0.2	1.0±0.1	1.1±0.2	1.0±0.1	1.1±0.1	1.2±0.2	1.2±0.1	1.2±0.1	1.05±0.08	1.1±0.1	1.04±0.08	1.06±0.06
^{230}Th/^{238}U	0.15±0.02	0.24±0.04	0.21±0.05	0.17±0.02	0.15±0.02	0.6±0.1	0.12±0.02	0.28±0.03	0.55±0.05	0.17±0.02	0.69±0.06	2.0±0.3	0.06±0.01	0.63±0.05	0.43±0.04	0.20±0.09	0.23±0.02	0.81±0.04
^{230}Th/^{234}U	0.11±0.02	0.18±0.03	0.16±0.03	0.13±0.02	0.12±0.02	0.38±0.07	0.09±0.01	0.27±0.03	0.52±0.07	0.18±0.02	0.61±0.05	1.7±0.2	0.05±0.01	0.54±0.04	0.41±0.04	0.18±0.06	0.22±0.02	0.76±0.04
^{226}Ra/^{230}Th	19±2	18±2	26±5	25±3	37±5	11±2	25±3	5.8±0.5	6.5±0.6	17±1	6.1±0.2	0.57±0.03	83±20	2.1±0.1	1.2±0.1	4.0±0.4	3.6±0.2	0.58±0.02
^{230}Th/^{232}Th	1.7±0.3	1.9±0.4	1.6±0.6	1.5±0.3	1.5±0.3	1.7±0.3	2.0±0.4	1.3±0.2	2.0±0.3	1.5±0.2	1.02±0.04	1.6±0.2	1.8±0.5	1.5±0.1	1.2±0.1	1.0±0.3	0.77±0.07	0.89±0.03
^{228}Th/^{232}Th	1.4±0.3	2.2±0.4	1.9±0.7	1.6±0.4	1.7±0.3	1.7±0.5	4.4±0.2	1.4±0.2	2.2±0.3	3.0±0.3	19.0±0.5	2.3±0.2	4±1	10.6±0.7	3.0±0.3	7.9±0.6	8.6±0.6	7.8±0.2
^{238}U/^{232}Th	11±2	8±1	8±3	9±2	10±2	3.0±0.7	17±3	4.4±0.5	3.70±0.06	9±1	1.5±0.1	0.8±0.1	28±6	2.3±0.2	2.7±0.3	5.0±0.5	3.44±0.03	1.09±0.05

TABLE 2: The Radioactivities of the Uranium and Thorium Family Members in the Hot Spring Waters and Sediments

Location code	P-1	P-2	P-3	P-4	H-1	H-2	M-1	L-1
Temp. (˚C)	98.0	79.5	72.3	70.5	79.0	51.0	26.0	71.0
pH	1.55	1.65	1.72	1.68	1.65	2.44	2.74	3.40
Hot Spring Water								
^{238}U (mBq/L)	85±4	67±2	68±2	76±3	14.4±0.7	4.9±0.4	2.6±0.2	2.3±0.2
^{234}U (mBq/L)	91±4	69±2	70±2	82±3	20.7±0.9	7.5±0.5	2.9±0.2	3.3±0.3
^{232}Th (mBq/L)	72±3	66±3	65±2	73±3	15.3±0.5	7.1±0.3	1.5±0.1	0.67±0.04
^{230}Th (mBq/L)	66±3	63±3	56±2	64±3	14.2±0.5	5.9±0.2	2.0±0.1	1.14±0.05
^{228}Th (mBq/L)	630±20	560±15	443±9	500±12	106±3	21.1±0.6	6.3±0.3	1.01±0.05
^{226}Ra (mBq/L)	36.5±0.7	29.0±0.8	N.D.	N.D.	8.96±0.05	7.2±0.1	10.8±0.1	4.5±0.1
^{234}U/^{238}U	1.1±0.1	1.03±0.06	1.03±0.06	1.08±0.08	1.4±0.1	1.5±0.2	1.1±0.2	1.4±0.3
^{230}Th/^{234}U	0.73±0.06	0.91±0.07	0.80±0.05	0.78±0.07	0.69±0.05	0.79±0.08	0.69±0.08	0.35±0.05
^{226}Ra/^{230}Th	0.55±0.04	0.46±0.03	—	—	0.63±0.03	1.22±0.06	5.4±0.3	3.9±0.3
^{228}Th/^{232}Th	8.8±0.6	8.5±0.6	6.8±0.3	6.8±0.4	6.9±0.4	3.0±0.2	4.2±0.5	1.5±0.2
Uranium (ppb)	6.6±0.3	5.4±0.1	5.5±0.2	6.1±0.2	1.16±0.06	0.40±0.03	0.21±0.02	0.18±0.02
Thorium (ppb)	17.8±0.9	16.4±0.8	16.1±0.6	18.0 ±0.7	3.8±0.1	1.75±0.07	0.36±0.03	0.17±0.01
U/Th (ppb/ppb)	0.37±0.04	0.33±0.02	0.34±0.03	0.34±0.02	0.31±0.02	0.23±0.03	0.6±0.1	1.1±0.2
Hot Spring Sediment								
^{238}U (Bq/kg (dry))			28.1±0.7	37±1		20.0±0.5		11.2±0.5
^{234}U (Bq/kg (dry))			29.6±0.7	38±1		20.6±0.5		11.3±0.5
^{232}Th (Bq/kg (dry))			57±4	66±4		18.8±0.9		15.2±0.7
^{230}Th (Bq/kg (dry))			36±3	54±4		16.0±0.8		15.0±0.7
^{228}Th (Bq/kg(dry))			750±30	1080±50		22±1		15.5±0.7
^{228}Ra (Bq/kg (dry))			2030±20	3010±30		31±1		18±1
^{226}Ra (Bq/kg (dry))			187±5	230±6		31±2		16±1
^{234}U/^{238}U			1.05±0.05	1.03±0.05		1.03±0.05		1.01±0.09
^{230}Th/^{234}U			1.2±0.1	1.4±0.1		0.78±0.06		1.3±0.1
^{226}Ra/^{230}Th			5.2±0.6	4.3±0.4		1.9±0.2		1.1±0.1
^{228}Ra/^{232}Th			36±3	46±3		1.6±0.1		1.2±0.1
^{228}Th/^{228}Ra			0.37±0.02	0.36±0.02		0.71±0.06		0.86±0.09
^{228}Th/^{232}Th			13±1	16±2		1.2±0.1		1.02±0.09
Uranium (ppm)			2.26±0.05	2.98±0.09		1.61±0.04		0.90±0.04
Thorium (ppm)			14.1±0.9	16 ±1		4.7±0.2		3.7±0.2
U/Th (ppm/ppm)			0.16±0.01	0.19±0.02		0.34±0.02		0.24±0.02

II. Hot Spring Water. The radioactivity concentrations of the hot spring water and its sediment samples in this study are listed in Tables 2 and 3, respectively. The temperatures and pH of the hot spring water vary greatly; their ranges are 26.0–98.0 ˚C and 1.55–3.40, respectively.

Radioactivity Distribution. As shown in Tables 2 and 3, the radioactivities of uranium, thorium and radium isotopes in hot spring water and sediments are distributed widely. The ranges of radioactivity in hot spring water for ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th and ^{226}Ra are 2.3–85, 2.9–91, 0.67–73, 1.14–66, 1.01–630, and 4.5–36.5 mBq/L, while those in hot spring sediments for ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{228}Ra and ^{226}Ra are 11.2–37, 11.3–38, 15.2–66, 15.0–54, 15.5–1080, 18–3010, and 16–230 Bq/kg (dry), respectively. Furthermore, the concentration ranges of uranium and thorium in hot spring water and sediments are 0.18–6.6, 0.17–18.0 ppb, and 0.90–2.98 ppm, respectively.

As shown in Figure 4 there is a reverse relationship of the concentration ratio of uranium/thorium vs. pH of hot spring water between hot spring water and sediment. Obviously, the concentration ratios of uranium/thorium in hot spring water are increased with the decrease of the acidity of hot spring water when pH > 2.5. Nevertheless, when pH < 2.5, the concentration ratios of uranium/thorium in hot spring water are slightly increased when the hot spring water becomes more acidic. And simultaneously, we can find the reverse relationship of

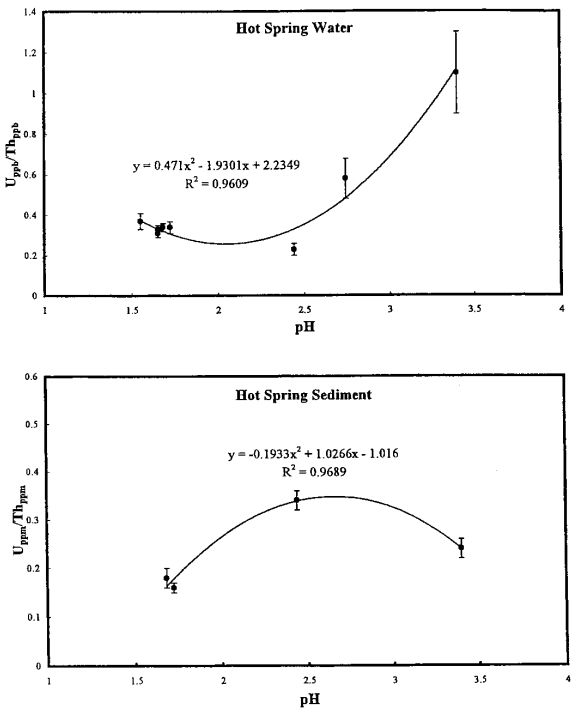

Figure 4. A plot of concentration ratios of U/ Th in hot spring water and sediments vs. pH of hot spring water.

concentration ratios of uranium/thorium vs. pH of hot spring water for hot spring sediments. As we have well known, uranium is mainly in the soluble ionic chemical forms of UO_2^{2+} in ground water at pH < 4.[10,11] However, thorium is more likely to be precipitated in the form of insoluble $Th(SO_4)_2^0$ when pH < 2.5, and mainly to become insoluble complexes with organic species when pH >2.5.[12] Therefore, it causes the higher concentration in thorium than in uranium in the sediments because of the formation of insoluble $Th(SO_4)_2^0$ or organic complexes of thorium at pH < 2.5 or pH > 2.5, respectively.

Radioactive Disequilibrium. The uranium and thorium series nuclides in hot spring water should come mainly from the leaching of the volcanic rocks. Leaching is a sorption/desorption process of the elements between the hot spring water and the rock. According to the age of the rock,[13] both uranium and thorium series nuclides in the rock should reach radioactive equilibrium, which means that the radioactivities of a radionuclide and its daughter nuclide are equal. If the radionuclide and its daughter nuclide are of the same isotopic elements, it means that they have the same chemical properties, their radioactivities should remain equal even after leaching. However, according to the results of Table 2, the radioactivity ratios of $^{234}U/^{238}U$ and $^{228}Th/^{232}Th$ were 1.03–1.5 and 1.5–8.8, respectively. Obviously, they are in radioactivity disequilibria.

1. $^{234}U/^{238}U$. As shown in Tables 2 and 3 the radioactivity radios of $^{234}U/^{238}U$ in hot spring water and sediments are 1.03–1.5 and 1.01–1.05, respectively. The radioactivity disequilibria for $^{234}U/^{238}U$ are not very serious in both hot spring water and sediments. It has been proven to be the result of α-recoil proceeding of ^{238}U decay, and the recoiled range of its daughter nuclide ^{234}Th ($T_{1/2}$ = 24.1 d) is about ~102 Å.[14] ^{234}Th recoiled atom can go through many lattice points and enter lattice defect pores or fractures of the rock and is easier to be leached.[15,16]

2. $^{228}Th/^{232}Th$. As it is known, just as ^{238}U and ^{234}U, ^{232}Th and ^{228}Th should be in radioactivity equilibrium in hot spring water and sediments because of the similar chemical properties. Figure 5, shows a diagram of radioactivity distributions of thorium series nuclides, i.e., ^{232}Th, ^{228}Ra, and ^{228}Th in hot spring water and sediments. It is obviously to be divided into two groups by pH < 2 and pH > 2. As shown in Figure 5, the radioactivities of ^{228}Th in both hot spring water and sediments and the radioactivities of ^{228}Ra in sediments are more enriched than ^{232}Th for the group of pH < 2. On the contrary, the radioactivities of ^{228}Th in both hot spring water and sediments and the radioactivities of ^{228}Ra in sediments are obviously equal to the same level of ^{232}Th for the group of pH > 2.

Furthermore, as listed in Table 2, the radioactivity ratios of $^{228}Th/^{232}Th$ in both hot spring water and sediments are increased from 1.5 to 8.8 and from 1.02 to 16, with the acidity of hot spring water from pH 3.40 to 1.55. There is obviously radioactivity disequilibrium between ^{232}Th and ^{228}Th. In the decay process from ^{232}Th to ^{228}Th, there is a middle nuclide ^{228}Ra with a long half-life ($T_{1/2}$ = 5.75 y). The chemical properties of radium and thorium are quite different in the aqueous solution. Therefore, we can assume that the enrichment of ^{228}Th may come from ^{228}Ra. To prove the above assumption, a diagram of the radioactivity ratios of $^{228}Ra/^{232}Th$ and $^{228}Th/^{228}Ra$ in sediments vs. pH of hot spring water is shown in Figure 6. As shown in Figure 6 we can find that ^{228}Ra is more enriched in sediments than ^{232}Th and ^{228}Th, and the radioactivity of ^{228}Ra in sediments is about 46 times and 2.8 times as high as that of ^{232}Th and ^{228}Th at pH = 1.68, respectively. It is the reason why the radioactivity of ^{228}Th is about 16 times as high as that of ^{232}Th in sediment at pH = 1.68, and leads the radioactivity of ^{228}Th to be about 9 times as high as that of ^{232}Th in hot spring water at pH = 1.55.

Conclusion

The hot spring and river waters at the Peitou hot spring basin in Northern Taiwan abounded with naturally occurring radionuclides. Generally, the radioactive concentrations of uranium-series and thorium-series nuclides in both river and hot spring waters were all pH dependent. Moreover, radioactive disequilibria were found both in $^{234}U/^{238}U$ and $^{228}Th/^{232}Th$. The enrichment of ^{228}Th to ^{232}Th is the most prominent. The excess of ^{228}Th comes from the contribution of ^{228}Ra.

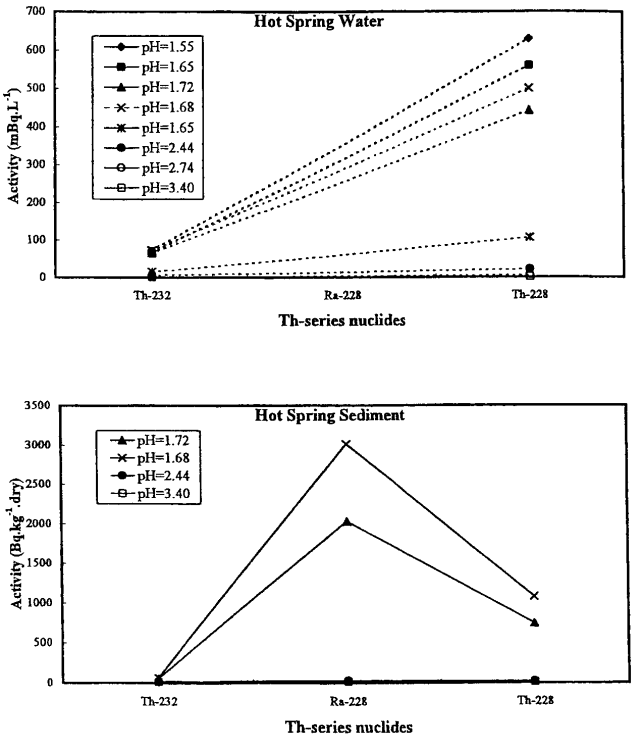

Figure 5. A plot of radioactivity of thorium series nuclides in hot spring water and sediments.

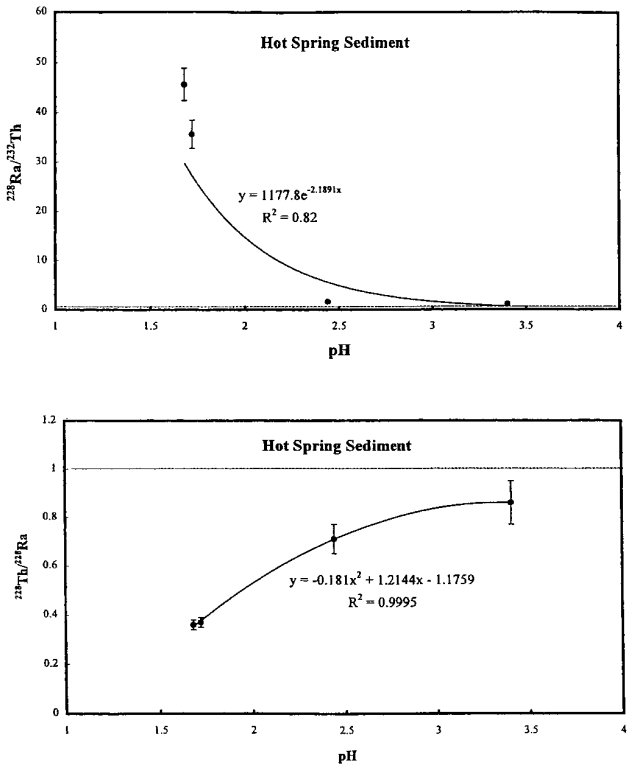

Figure 6. A plot of radioactivity ratios of $^{228}Ra/^{232}Th$ and $^{228}Th/^{228}Ra$ in hot spring sediments vs. pH of hot spring water.

References

(1) Y. Okamoto, J. Geol. Soc. Tokyo **16**, 72 (1909).
(2) Y. Okamoto, J. Geol. Soc. Tokyo **18**, 19 (1911).
(3) F. H. Chang, Acta Geol. Taiwanica **9**, 7 (1961).
(4) H. Hamaguchi, Y. T. Lee, and H. S. Cheng, J. Chin. Chem. Soc. **9**, 1 (1962).
(5) P. S. Weng and C. M. Tsai, Health Phys. **24**, 429 (1973).
(6) Science and Technology Agency in Japan, *Analytical Methods of Uranium* (in Japanese) (Japan Chemical Analysis Center, Chiba, Japan, 1987).
(7) F. S. Jiang, S. C. Lee, S. N. Baktiar, and P. K. Kuroda, J. Radioanal. Nucl. Chem., Articles **100**, 65 (1986).
(8) Science and Technology Agency in Japan, *Analytical Methods of Radium* (in Japanese) (Japan Chemical Analysis Center, Chiba, Japan, 1990).
(9) R. M. Smith and A. E. Martell, *Critical stability constants* (Plenum press, New York and London, 1976) p.321.
(10) D. Langmuir, Geochim. et Cosmochim. Acta **42**, 547 (1978).
(11) J. S. Tripathi, Geochim. et Cosmochim. Acta **43**, 1989 (1979).
(12) D. Langmuir, Geochim. et Cosmochim. Acta **44**, 1753 (1980).
(13) W. S. Juang, *Geochronology and geochemistry of late cenozoic volcanic rocks from Taiwan*, Ph. D. Thesis (in Chinese) (Oceanogr., National Taiwan University, Taipei, 1988) p.19.
(14) K. Kigoshi, Science **173**, 47 (1971).
(15) J. N. Rosholt, W. R. Shields, and E. L. Garner, Science **139**, 224 (1963).
(16) I. O. Essien, J. Radioanal. Nucl. Chem. **139**, 331 (1990).