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Geochemical significance of ^{14}C, ^{3}H, δ^{18}O, δ^{2}H and ^{87}Sr/^{86}Sr isotope data for the Dongrae and Haeundae hot spring waters, Busan, South Korea

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ABSTRACT: Dongrae and Haeundae are representative hot spring areas that have been used as spas for more than 1,000 years in the southern Korean Peninsula. These hot springs have water temperatures >58 °C and are located along the southeast coastal area of the peninsula. We used ^{14}C, ^{3}H, δ^{18}O, δ^{2}H, and ^{87}Sr/^{86}Sr isotope data for the Dongrae and Haeundae hot spring waters collected over 2004–2014 to investigate the groundwater cycle and heat source for these hot springs. The stable isotope compositions of O and H suggested meteoric origin of the hot spring waters. The ^{87}Sr/^{86}Sr ratios for the Dongrae and Haeundae hot spring waters have been steady for 11 years at 0.70567 ± 0.00002 and 0.70607 ± 0.00002, respectively, suggesting that they are in a near equilibrium state. The ^{14}C age of the Dongrae hot spring waters ranges from 1,401 to 2979 years BP, and that of the Haeundae hot spring waters from 1930 to 6687 years BP. We observed a strong correlation between ^{87}Sr/^{86}Sr and ^{14}C, as well as δ^{18}O. Therefore, the hot springs in the Busan area were supposed to be heated by a paleo-heat source, suggesting that there may be no current heat source under the present crustal conditions. This study also demonstrates that monitoring of the ^{87}Sr/^{86}Sr ratio and ^{14}C in the groundwater and deep thermal water can be used as a proxy for tracing the heat source of geothermal waters such as hot springs and an indicator of groundwater mixings between upper and lower aquifers in granite area.

Key words: hot springs, ^{87}Sr/^{86}Sr, ^{14}C, groundwater mixing

1. INTRODUTION

Hot springs are produced by emergence of geothermally heated groundwater from the earth's crust. Most hot springs are located in areas with volcanic activity or active faults as a result of eruptive events or persistent hydrothermal systems (Graham, 1992; Vengosh et al., 2002; Möller et al., 2004; Du et al., 2005; Papp and Nitoi, 2006; Sanada et al., 2006). However, despite being a non-volcanic area, the Korean Peninsula has 14 hot spring areas with temperatures >40 °C. The presence of these hot springs is related to the distribution of Mesozoic granites over about one-third of the southern Korean Peninsula with a bimodal age distribution, i.e., Jurassic and Cretaceous (Lee et al., 2011). Lee et al. (2011) reported that the ^{87}Sr/^{86}Sr ratio of the hot spring waters of South Korea was closely associated with the distribution of Mesozoic granite.

Dongrae and Haeundae in the Busan city are representative areas with hot springs in South Korea, and the hot springs have been used as spas for more than 1,000 years. The two hot-spring areas are located in diagonal direction along the large Dongrae fault (Fig. 1). Recently, Lee et al. (2008, 2009) suggested that, based on ^{87}Sr/^{86}Sr ratio and ^{14}C age of the Dongrae hot spring water, it may originate from a high-temperature paleo-groundwater reservoir rather than from circulation of current meteoric water. However, there has been no previous comparison of the isotope geochemistry between the Dongrae and the Haeundae hot springs for clarifying their water and heat source in the Busan area.

We used chemical composition data as well as ^{14}C, ^{3}H, δ^{18}O, δ^{2}H, and ^{87}Sr/^{86}Sr isotope data for the Dongrae and Haeundae hot spring waters collected over the last 10 years to discuss: 1) the origin of the water for each hot spring, 2) the relationship between the Dongrae and Haeundae hot spring waters, and 3) the heat source for the hot spring waters in the Busan area. Our results provide a framework for better understanding the heat sources of high-temperature deep groundwater such as hot spring waters in old granite areas. These results may also play an important role in sustainable utilization and protection of geothermal waters in non-volcanic areas.

2. GEOLOGIC SETTING AND HYDROGEOLOGIC CHARACTERISITCS OF THE HOT SPRINGS

2.1. Geologic Setting

The Dongrae and Haeundae hot springs are located at the southeast margin of the Korean Peninsula in the Cretaceous granitoid area (Fig. 1). The granites of the Dongrae hot spring area were crystallized from a calc-alkaline series, and I-type granitic rocks that evolved from granodioritic magma into hornblende granite, adamellite, biotite granite, and finally

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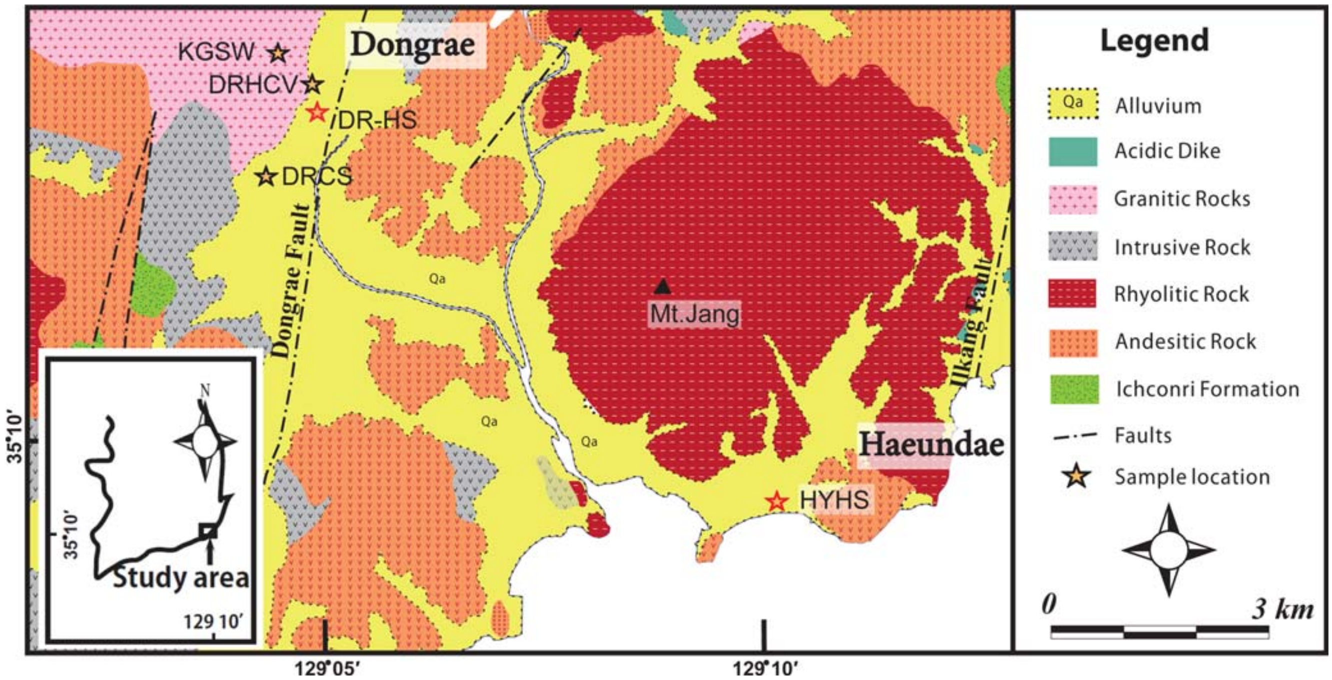

Fig. 1. Geologic and sample location map. The boreholes for the hot spring waters (for example DRHS and DRHSHS) in the Dongrae area are located within diameter 200 m. KGSW is the sampling location of the surface water, and DRCS is the sampling location of shallow groundwater. The distance between HYHS and HYOHS in Haeundae area is within 500 m.

micrographic granite through fractional crystallization of plagioclase (Yun et al., 2005). Yun et al. (2005) also proposed, based on trace element composition and rare earth element (REE) patterns, that the granitoids were continental margin arc calc-alkaline rocks produced in a subduction environment at 69.6 ± 1.9 Ma (2σ) with an initial $^{87}Sr/^{86}Sr$ ratio of 0.70503 ± 0.00015.

The felsic volcanic–plutonic sequence in the Haeundae area can be divided into early rhyolite, rhyolitic ash flow tuff, late rhyolite, granite porphyry, and feldspar porphyry. Yun et al. (2005) also suggested that the emplacement pressure of the rhyolitic ash as flow tuff can be divided into two groups, 4.0–5.4 kbar and 1.5–2.6 kbar, and that equilibrium temperatures of the rhyolitic magma chamber based on plagioclase-hornblende geothermometer were 845 and 737 °C.

2.2. Location and Hydrology

The Dongrae hot springs originate near the Dongrae fault line. Son et al. (2002) suggested that the topographical basin formed at Sanseong town, Geumjeong Mt. (north of the sampling site for KGSW in Fig. 1) may be a major recharge area for groundwater in the study area. The authors also argued that the recharged groundwater in the basin might be circulated to the depth of about 3–4 km and finally would reach the Dongrae hot spring area. In addition, Ryu et al. (2007) proposed that the shallow groundwater in the Busan area flows towards between south and southwest. These suggest that groundwater flows from the western mountain range to the lower elevation area of southeastern Dongrae.

The Haeundae hot spring is located about 15 km southeast of the Dongrae hot spring at the edge of the Cretaceous andesitic rock. Shim et al. (2000) reported that the thermal water likely flows along the fracture in a 30°N–60°E direction with dip of 60–80°SE or 60–80°NW.

3. SAMPLES AND METHODS

3.1. Samples

Water samples from the Dongrae and Haeundae hot springs (DRHS and HYHS, respectively) have been collected annually in 2004–2014 (Tables 1 and 2). Additional hot spring water samples collected in the Dongrae area during 2007–2011 (e.g., DRHSHS-A, DRHSHS-B, DRNCHS, and DRGCHS) were selected for comparison with DRHS, and HYOCHS in Haeundae area was selected for comparison with HYHS at 2010. The hot springs in each area are located within ~500 m of each other. Water samples were filtered through 0.45 μm millipore filters and divided into two parts. One subsample was used for anion analysis, and the other was acidified with double-distilled HNO_3 and sealed in 100- and 250-mL pre-cleaned polyethylene bottles for Sr isotope and cation analysis, respectively. Unacidified water samples were also collected in 2 100-mL pre-cleaned polyethylene bottles for anion determination and stable isotope analysis including

Table 1. Sr isotopic composition of the thermal water, groundwater and rainwater at Dongrae area, Korea

Sample name		Sampling date	well depth (m)	T (°C)	pH	EC (μS/cm)	$^{87}Sr/^{86}Sr$	2σ	Sr (μg/L)	
DRHS-01	thermal water	Feb., 2004	220	61	8.34	1542	0.70569	0.00001	1060	Lee et al. (2008)
DRHS-02		Aug., 2004	220	57.7	7.48	1472	0.70569	0.00001	883	Lee et al. (2008)
DRHS-03		Mar., 2005	220	61	7.85	1540	0.70569	0.00001	902	Lee et al. (2008)
DRHS-04		Mar., 2006	220	62.8	8.02	1463	0.70569	0.00001	1253	Lee et al. (2008)
DRHS-05		Mar., 2007	220	63.4	7.93	1551	0.70567	0.00001	1080	Lee et al. (2008)
DRHS-06		Jun., 2007	220	63	7.9	1521	0.70568	0.00001	1080	Lee et al. (2008)
DRHS-07		Nov., 2007	220	62.1	7.92	1515	0.70570	0.00001	1117	Lee et al. (2008)
DRHS-08		Mar., 2008	220	63	7.9	1521	0.70566	0.00001	1100	Lee et al. (2009)
DRHS-08-02		Dec., 2008	220	62	8.2	1485	0.70569	0.00001	960	Lee et al. (2009)
DRHS-09		Apr., 2009	220	62.7	8.02	1570	0.70569	0.00001	1031	this study
DRHS-10		Apr., 2010	220	62	7.68	1473	0.70568	0.00002	922	this study
DRHS-11		Apr., 2011	220	64.5	8.06	1876	0.70568	0.00001	1006	this study
DRHS-12		May, 2012	220	65.2	8.02	1563	0.70569	0.00001	1075	this study
DRHS-13		Jul., 2013	220	67.1	8.19	2100	0.70568	0.00001	1170	this study
DRHSHS-A-05		Mar., 2007	175	49.5	7.44	1158	0.70567	0.00001	1110	Lee et al. (2008)
DRHSHS-A-08		Jun., 2008	175	–	–	–	0.70563	0.00001	998	Lee et al. (2009)
DRHSHS--09		Apr., 2009	175	50	7.57	1095	0.70567	0.00001	758	this study
DRHSHS-A-10		May, 2010	175	48	7.4	1128	0.70565	0.00001	801	this study
DRHSHS-A-11		Apr., 2011	175	50.4	7.74	1238	0.70566	0.00001	697	this study
DRHSHS-B-05		Mar., 2007	230	58	7.68	1385	0.70565	0.00001	880	Lee et al. (2008)
DRHSHS-B-08		Jun., 2008	230	–	–	–	0.70572	0.00001	828	Lee et al. (2009)
DRHSHS-B-10		May, 2010	230	52	7.79	1473	0.70566	0.00001	899	this study
DRHSHS--11		Apr., 2011	230	59.4	7.8	1418	0.70567	0.00001	905	this study
DRGCHS-10		Jun., 2010	210	62.8	7.77	1501	0.70568	0.00001	1150	this study
DRNCHS-10		Jun., 2010	210	68.8	8.03	1731	0.70566	0.00001	982	this study
DRNCHS-14		Jul., 2014	210	60.1	7.75	1451	0.70569	0.00001	1170	this study
DRHCV	groundwater	Jun., 2010	150	20	7.03	970	0.70561	0.00001	409	this study
DRCS-01		Mar., 2007	130	8.6	7.31	623	0.70578	0.00003	384	Lee et al. (2008)
DRCS-02		Nov., 2007	130	16.4	7.13	630	0.70579	0.00001	408	Lee et al. (2008)
DRGV		Jun., 2008	–	–	–	–	0.70629	0.00001	86.6	Lee et al. (2009)
KGSW	surface water	Nov., 2007	–	13.5	7.3	69.4	0.70670	0.00001	30	Lee et al. (2008)
GeumJeongSan		Jun., 2010	30	17	8	36	0.70741	0.00001	5	this study
DRP20060630	rain water	Jun., 2006	–	–	–	–	0.71235	0.00001	4.8	this study
DRP20070625	rain water	Jun., 2007	–	–	–	–	0.70738	0.00001	1	Lee et al. (2008)

δD (δ^2H), $\delta^{18}O$, and ^{13}C. A 500-mL pre-cleaned polyethylene bottle was also used for collection of a ^{14}C water sample.

3.2. Analytical Methods

Sr concentrations were analyzed by the inductively coupled plasma atomic emission spectroscopy (ICP-AES) at the Korea Institute of Geoscience and Mineral Resources (KIGAM), Daejeon, Korea. The oxygen ($\delta^{18}O$), hydrogen (δD), and carbon (^{13}C) isotopes were analyzed by the stable isotope ratio mass spectrometry using a Micromass Optima mass spectrometer for $\delta^{18}O$ and ^{13}C, and a GV Instruments Iso Prime Micromass spectrometer for δD at the Korean Basic Science Institute (KBSI), Ochang, Korea. For ^{18}O analysis, about 0.2 mL of the water sample was equilibrated with CO_2 gas at 25.0 ± 0.1 °C (Epstein and Mayeda, 1953). The CO_2 gas was then extracted and cryogenically purified. For D analysis, Cr metal was used to produce hydrogen gas using an automatic on-line sample preparation system (Pyr-OH Model, GV Instruments) (Morrison et al., 2001). Dissolved inorganic carbon (DIC) was then separated for ^{13}C isotope analysis following Atekwana and Krishnamurthy (1998). The $\delta^{18}O$, δD, and ^{13}C were referenced to Vienna Standard Mean Ocean Water (VSMOW) and Vienna-Pee Dee Belemnite (VPDB) with a precision of ±0.1‰, ±1.0‰, and 0.2‰, respectively.

DIC in the hot spring water and groundwater was pre-

Table 2. Sr isotopic composition of the hot spring water at Haeundae area, Busan, Korea

Sample name	Sampling date	well depth (m)	T (ºC)	pH	EC (μS/cm)	$^{87}Sr/^{86}Sr$	2σ	Sr (μg/L)	
HYHS-01	Feb., 2004	800	56.9	7.49	6,710	0.70607	0.00001	7160	this study
HYHS-02	Aug., 2004	800	57.4	7.17	6,770	0.70606	0.00001	7230	this study
HYHS-05	Mar., 2005	800	57.4	7.75	6,690	0.70608	0.00001	7240	this study
HYHS-06	Mar., 2006	800	56.8	7.38	6,850	0.70608	0.00001	6920	this study
HYHS-07	Mar., 2007	800	57.0	7.46	6,380	0.70608	0.00001	6800	this study
HYHS-08-1	Mar., 2008	800	56.0	7.06	6,350	0.70608	0.00001	6640	this study
HYHS-08-2	Nov., 2008	800	56.0	7.67	6,630	0.70607	0.00001	6748	this study
HYHS-09	Apr., 2009	800	54.0	7.59	6,890	0.70602	0.00001	6940	this study
HYHS-10	Apr., 2010	800	56.0	7.27	6,410	0.70606	0.00001	6291	this study
HYHS-11	Apr., 2011	800	56.5	7.5	6,480	0.70605	0.00001	6440	this study
HYHS-12	Apr., 2012	800	56.8	7.36	6,370	0.70607	0.00001	6677	this study
HYHS-13	Jul., 2013	800	58.5	7.48	6,510	0.70607	0.00001	6690	this study
HYHS-14	Jul, 2013	800	56.9	7.07	6,340	0.70606	0.00001	6690	this study
HYOCHS-10	Jun., 2010	–	54.5	7.47	7,540	0.70561	0.00001	6810	this study

cipitated as $SrCO_3$ by adding $SrCl_2$ in NH_4OH solution to the water samples in the laboratory. The precipitated $SrCO_3$ was converted to CO_2 by reacting it with phosphoric acid in a vacuum line at Nagoya University, Japan. The resulting CO_2 was collected and purified in a vacuum line and subsequently reduced to graphite with an iron catalyst and hydrogen at 650 °C for 6 h (Watanabe et al., 2013). The ^{14}C analysis was performed with the Tandetron Accelerator Mass Spectrometry System (AMS, Model-4130, High Voltage Engineering Europe) at the Center for Chronological Research, Nagoya University, Japan. The relative standard deviations of the $^{14}C/^{12}C$ ratios were generally ±0.3–0.5%. The HOx-II standard (oxalic acid, SRM-4990C; National Institute of Standard and Technology) was used as the ^{14}C concentration reference.

The $^{87}Sr/^{86}Sr$ ratio was measured using a VG Sector 54-30 multi-collector thermal ionization mass spectrometer at KBSI, except for samples DRHS-13, DRNCH-14, HYHS-13 and HYHS-14. The $^{87}Sr/^{86}Sr$ ratio in samples DRHS-13 DRNCH-14, HYHS-13 and HYHS -14 was measured using a TRITON TIMS at KIGAM. The average value for NBS 987 during the analyses in KBSI and KIGAM were 710244 ± 0.000004 (2 σ_m) for 30 measurements and 710249 ± 0.000002 (2 σ_m) for 30 measurements, respectively. The analytical uncertainty was approximately 0.002% for $^{87}Sr/^{86}Sr$. The total concentrations in procedural blanks during the Sr isotopic measurements were <30 pg.

4. RESULTS

4.1. Water Chemistry

The geochemical characteristics of the Dongrae and Haeundae hot spring waters have been reported previously (Han et al., 1999; Shim et al., 2000; Sung et al., 2001; Lee et al., 2008, 2009). Lee et al. (2008) showed that, in the Piper diagram, the Dongrae hot spring water plotted in the area of the Na-Cl type whereas the cold groundwater plotted in the area of the $Ca\text{-}HCO_3$ type. Shim et al. (2000) reported that the groundwater in the Haeundae area belonged $Ca\text{-}HCO_3$ type, while the hot spring water belonged to Na-Cl type. The Dongrae hot spring water and cold groundwater differed in electrical conductivity (EC) and pH. The EC of the Dongrae hot spring water was 1,095–2,100 μS/cm. However, EC of shallow groundwater is lower than 1,000 μS/cm. The pH of the hot spring water was 7.68–8.39, whereas that of the shallow groundwater was 7.03–7.31 (Table 1). The EC of the Haeundae hot spring water was 6,350–6,890 μS/cm, while the EC of the nearby hot spring water, HYOCHS-10, was 7,450 μS/cm, slightly higher than that of HYHS (Table 2). The water temperature and pH of the two hot spring waters were similar and both were the Na-Cl type. This geochemical characteristic is similar to those of Shim et al. (2000).

Figure 2 is a piper diagram for the hot spring water, groundwater, surface water and seawater near to the Dongare and Haeundae areas. The figure was plotted based on the data from the previous literatures. Figure 2 represents well that the hydrogeochemical characteristics of the Dongrae and Haeundae hot spring waters belong to Na-Cl type, however they are different from those of groundwater and surface water in the neighboring area. Figure 2 also indicates that the Haeundae hot spring water apparently has more stronger chemical composition of sea water than the Dongrae hot spring water. However, hydrogeochemical characteristics of the hot spring waters from these two areas are different from that of sea water for Mg contents. The Mg contents of the two hot spring waters are much lower than that of sea water. This may indicate that the two hot spring waters are not affected by current sea water.

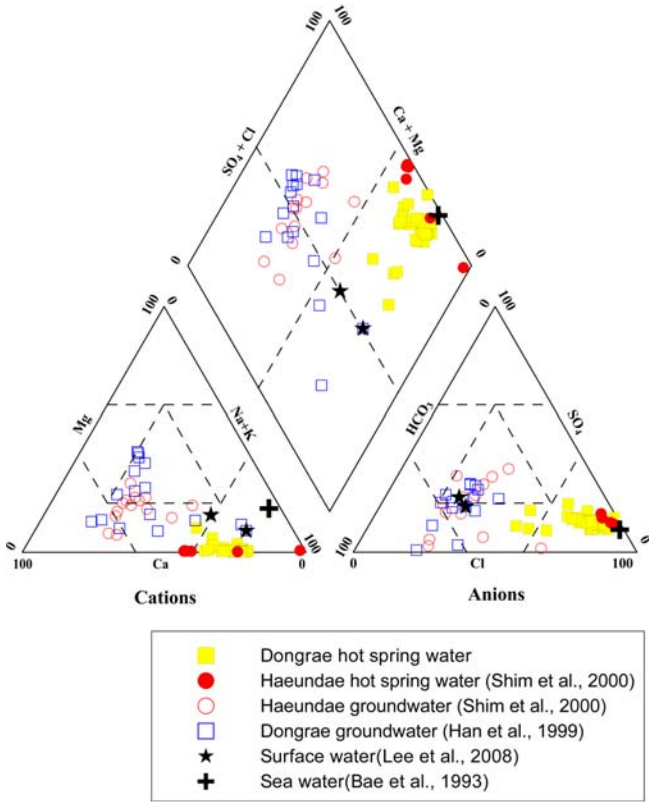

Fig. 2. Piper diagram of the hot spring waters, groundwater, surface water, and sea-water in the Busan area, South Korea (modified from Lee et al., 2008, 2009). The data of the Dongrae hot spring waters are from Han et al. (1999), Sung et al. (2001), and Lee et al. (2008).

4.2. $\delta^{18}O$, δD, ^{13}C, and ^{14}C Isotope Data

The δD, $\delta^{18}O$, ^{13}C, and ^{14}C isotope data are shown in Table 3. The δD values for the hot spring waters ranged from –58.0‰ to –52.5‰, and the $\delta^{18}O$ values ranged from –9.33‰ to –7.71‰. Therefore, the hot spring waters in the Dongrae and Haeundae areas are of meteoric origin, plotting along the global meteoric water line (GMWL; Fig. 3).

The ^{3}H concentrations for DRHS and HYHS were 0.78–0.88 TU and 0.54–1.74 TU, respectively, during 2009–2010. However, since 2012, ^{3}H has not been detected in the hot spring waters. The detection of ^{3}H concentrations during 2009–2010 seems to be due to some contamination during sampling or unskilled sample handling. The $\delta^{13}C$ values for DIC from the Dongrae hot spring water ranged from –19.5‰ to –15.7‰, whereas those from the Haeundae hot spring water ranged from –16.6‰ to –10.5‰. The $\delta^{13}C$ values of DIC from the shallow groundwater in the Dongrae area ranged from –20.5‰ to –14.4‰. Measured ^{14}C concentrations for the Dongrae hot spring waters were 69–84 pMC whereas those for the Haeundae hot spring waters were 43.5–78.6 pMC. However, δD, $\delta^{18}O$, and ^{14}C values in water from the site Hanshin CityVil in the Dongrae area (sample DRHCV), a mixture of hot spring water and cold shallow groundwater, were –50.2‰, –7.02‰, and 99.7 pMC, respectively.

4.3. $^{87}Sr/^{86}Sr$ Ratios and Strontium Concentrations

The Sr isotope and concentration data for Dongrae and Haeundae during 2004–2014 are shown in the Tables 1 and 2, respectively. Sr concentrations in Dongrae hot spring waters were 697–1,253 μg/L. In contrast, shallow cold groundwater and surface water had Sr concentrations of 5–409 μg/L. The Sr concentrations in Haeundae hot spring waters were 6,291–7,240 μg/L. By comparison, rain water has Sr concentrations ranging from 1–4.8 μg/L (Lee et al., 2011).

Figures 4a and 5a show $^{87}Sr/^{86}Sr$ ratio for the Dongrae and Haeundae hot spring waters during 2004–2014, respectively. The $^{87}Sr/^{86}Sr$ ratios for both Dongrae and Haeundae hot spring waters were nearly constant, except for HYOCHS from a Haeundae hot spring. The $^{87}Sr/^{86}Sr$ ratios for hot spring waters from the Dongrae area were 0.70563–0.70569 (mean, 0.70568 ± 0.00002, 1σ), except for DRHSHS-B-08 (0.70572). Those for Haeundae were 0.70602–0.70608 (mean, 0.70607 ± 0.00002, 1σ), except for HYOCHS (0.70561). The $^{87}Sr/^{86}Sr$ ratios for the cold groundwater, surface water and rain water were in the range 0.70578–0.71235, except for DRHCV (0.70561), which was a mixture of groundwater and hot spring water. The $^{87}Sr/^{86}Sr$ ratios for the cold groundwater were slightly higher than those for the Dongrae hot spring waters, and the $^{87}Sr/^{86}Sr$ ratios for surface water and rain-water were higher than those of any of the hot spring waters in the Busan area.

5. DISCUSSION

5.1. δD and $\delta^{18}O$ Isotope Constraints on the Origin of the Hot Spring Water

The water source is an important factor to consider in determining the heat source for the hot springs. Sung et al. (2001) proposed that the hot springs in southern South Korea originated from mixing between high-temperature deep groundwater and present sea water, along with seawater-rock interaction. However, based on isotope data, e.g., ^{14}C and $^{87}Sr/^{86}Sr$ ratios for hot spring water, shallow groundwater, surface water, and rainfall, Lee et al. (2008, 2009) suggested that the geochemical characteristics of the sea-water in DRHS indicate remnants of past sea-water rather than present sea-water mixing.

The δD and $\delta^{18}O$ isotopic composition of water can provide information on recharge patterns of groundwater including recharge altitude and water sources (Katz et al., 1998; Abbott et al., 2000). Lee and Lee (1999) summarized that there are two kinds of local meteoric water lines (LMWL) in the Korean Peninsula according to rainy season (summer) and dry season (winter). The LMWL during summer season was plotted on the line of GMWL by Craig (1961). Recently, Koh et al. (2010)

Table 3. Isotope composition of the hot spring water and shallow cold groundwater in the Dongrae and Haeundae areas

Area	Sample	Kind[a]	δD (‰SMOW)	δ^{18}O (‰SMOW)	δ^{13}C[b] (‰)	^{3}H (TU)	^{14}C conc.[c] (pMC, %)	(±) 1σ ^{14}C conc.	^{14}C age (yr BP)	(±) 1σ^{14}C age	
Dongrae	DRHS-08-01	H	–	–	–19.5	–	81.0	0.5	1692	37	Lee et al. (2009)
	DRHS-08-1-re	H	–	–	–18.3	–	81.5	0.4	1641	32	this study
	DRHS-09	H	–54.9	–8.13	–17.9	0.78 ± 0.09	79.4	0.4	1855	32	this study
	DRHS-10	H	–55.4	–8.02	–15.7	0.88 ± 0.76	84.0	0.3	1402	26	this study
	DRHS-11	H	–55.5	–8.15	–18.4	–	76.6	0.3	2136	27	this study
	DRHS-12	H	–56.6	–8.05	–18.3	<0.5	–	–	–	–	this study
	DRHS-13	H	–58	–8.14	–18.1	<0.5	–	–	–	–	this study
	DRHSHS-A-08	H	–52.5	–7.75	–18.1	–	85.4	0.4	1271	36	Lee et al. (2009)
	DRHSHS-A-10	H	–54.2	–7.78	–17	–	80.5	0.0	1746	25	this study
	DRHSHS-A-11	H	–53.0	–7.71	–18.3	–	77.8	0.3	2021	27	this study
	DRHSHS-B-08	H	–	–	–19.2	–	73.6	0.5	2467	36	Lee et al. (2009)
	DRHSHS-B-09	H	–	–	–18.4	–	75.8	0.4	2227	32	this study
	DRHSHS-B-10	H	–54.1	–7.91	–17.6	–	70.1	0.3	2852	27	this study
	DRHSHS-B-11	H	–54.6	–7.96	–19.2	–	69.0	0.2	2979	29	this study
	DRNCHS-10	H	–	–	–17.0	–	82.5	0.3	1544	21	this study
	DRNHS-14	H	–54.1	–8.14	–17.8	<0.5	–	–	–	–	this study
	DRGCHS-10-1	H	–55.5	–9.33	–16.7	–	72.2	0.3	2609	21	this study
	DRGCHS-10-2	H	–55.1	–8.05	–19.9	–	71.0	0.3	2748	22	this study
	DRHCV	C	–50.2	–7.02	–16.0	–	94.8	0.3	430	23	this study
	DRGV	C	–	–	–20.5	–	106.4	0.4	–495	33	Lee et al. (2009)
	DRGV-07-2	C	–	–	–19.5	–	99.7	0.4	25	30	this study
	GeumJeongSan	C	–	–	–14.4	–	–	–	–	–	this study
Haeundae	HYHS-09	H	–53.4	–7.89	–	0.54 ± 0.07	71.3	0.4	2714	33	this study
	HYHS-10	H	–54.3	–7.86	–15.6	1.74 ± 0.75	77.6	0.3	2037	26	this study
	HYHS-11	H	–53.9	–7.85	–16.6	–	78.6	0.5	1930	50	this study
	HYHS-12	H	–55.5	–7.89	–15.4	<0.5	–	–	–	–	this study
	HYHS-13	H	–54.8	–7.81	–16.5	<0.5	74.9	0.5	2320	50	this study
	HYHS-14	H	–51.9	–7.87	–15.7	<0.5	–	–	–	–	this study
	HYOCHS-10	H	–53.5	–7.85	–14.8	–	43.5	0.5	6687	42	this study

[a] H: hot spring water, C: shallow cold groundwater.

[b] $\delta^{13}C = [(^{13}C/^{12}C)_{sample}/(^{13}C/^{12}C)_{PDB} - 1.0] \times 1000$ (‰), PDB means Pee Dee Belemnite which is a standard material from a fossil.

[c] Except HYHS-13, ^{14}C concentration of the water samples was measured using TADETRON-AMS at Nagoya University.

pMC: present Modern Carbon.

^{14}C concentration of HYHS-13 was measured by 41000Bo-AMS (High Voltage Engineering Europa) at KIGAM.

mentioned that the granitic aquifers in mainland Korea are recharged mainly by summer precipitation due to much higher rainfall intensity in summer.

Figure 3 shows that hot spring waters from the two areas plot along the LMWL during summer season by Lee and Lee (1999) except for DRGCHS10. DRGHS10 falls on the line of LMWL during winter season. However, surface water and sea-water deviate from the GMWL. The absence of a recognizable oxygen isotope shift in Figure 3 appears to be related to the relatively low reservoir temperature (Taylor, 1974; Yun et al., 1998). These geochemical characteristics of the Dongrae and Haeundae hot spring waters suggest that they were mainly derived from meteoric water during summer season. Therefore, we may be able to assume that the present hot spring waters originate from geothermal waters in deep circulation under a deep geothermal reservoir.

5.2. Geochemical Implication of Strontium Concentrations and ^{87}Sr/^{86}Sr Ratios

The Sr concentrations in the Dongrae and Haeundae hot spring waters were relatively high and variable over the last 11 years compared with neighboring groundwater and surface water (Tables 1 and 2). The primary sources of Sr in groundwater are meteoric inputs and dissolution of Sr-bearing minerals. The chemical behavior of Sr in groundwater

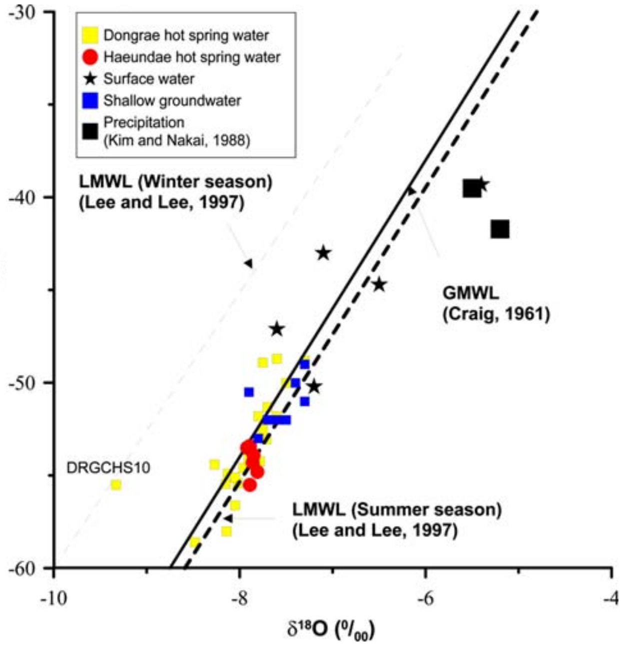

Fig. 3. Isotope composition (δD and δ^{18}O) of the hot spring waters, groundwater, surface water, and sea-water in the Busan area, South Korea. LMWL and GMWL represent the local meteoric water line in South Korea (Lee and Chung, 1997; Lee and Lee, 1999) and the global meteoric water line (Craig, 1961), respectively.

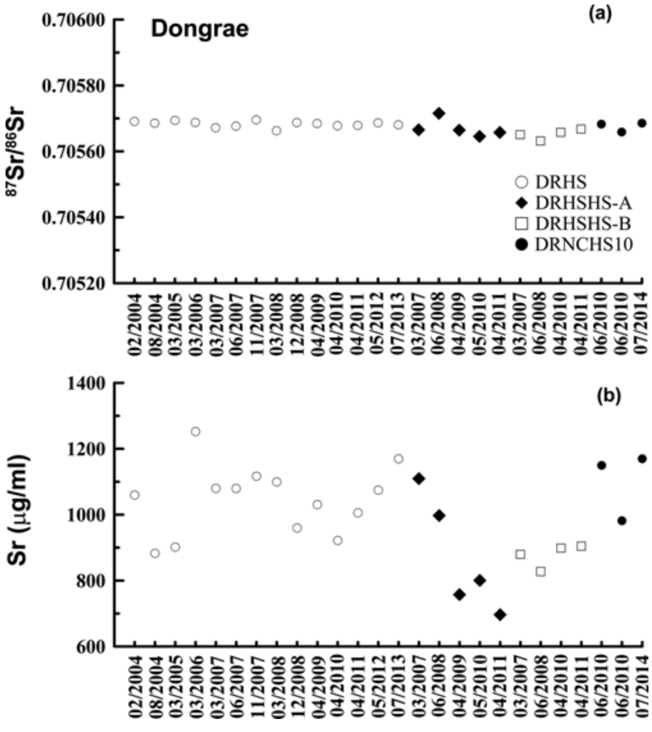

Fig. 4. Variations in the (a) ^{87}Sr/^{86}Sr ratio and (b) Sr concentration of the hot spring water during 2004–2013 in the Dongrae area.

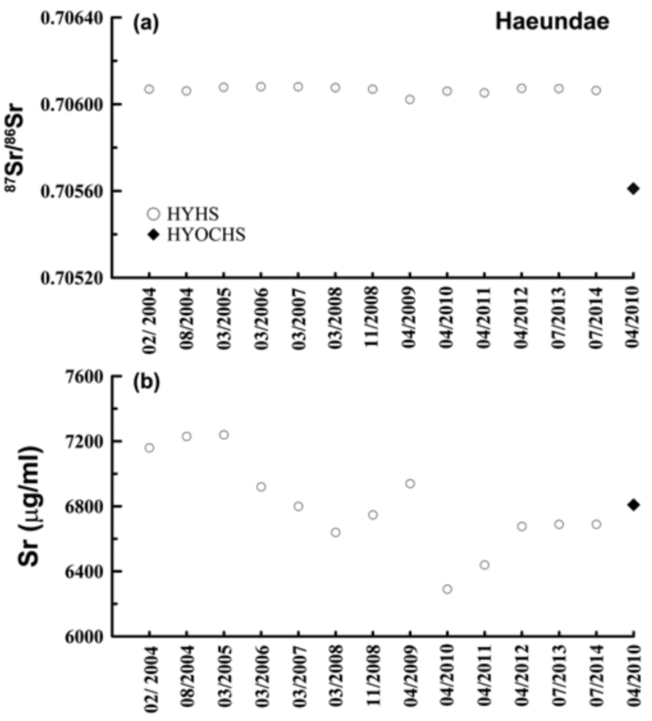

Fig. 5. Variations in the (a) ^{87}Sr/^{86}Sr ratio and (b) Sr concentration of the hot spring water during 2004–2013 in the Haeundae area.

is affected by various parameters, including the water temperature, the aquifer-bearing rock type, and minerals such as plagioclase and calcite. Négrel and Deschamps (1996) and Négrel et al. (2001) also proposed the possibility of anthropogenic influences such as fertilizers and deicing salts on groundwaters. Recently, Lee et al. (2011) concluded that any anthropogenic input of Sr to hot spring waters in South Korea is likely negligible, because most hot spring waters are directly pumped through pipes protected by casing from a depth of 70–460 m below bedrock to prevent infiltration by cold shallow groundwater. The Sr concentrations in the Dongrae and Haeundae hot spring waters have not been constant over 10 years, which may be due to water-rock interactions or mixing with sea-water (Figs. 4b and 5b).

The ^{87}Sr/^{86}Sr ratios for the Dongrae and Haeundae hot spring waters in the Busan area have been stable over last 11 years (Figs. 4a and 5a). Therefore, variations in such a stable ^{87}Sr/^{86}Sr ratio would not be expected in the near future. The ^{87}Sr/^{86}Sr ratios for rain water in South Korea are 0.7074–0.7128 (Jeon and Chung, 2005; Lee et al., 2008), which are higher than those of the hot spring water in the Busan area. Lee and Kim (2010) demonstrated during a 12-month long laboratory experiment that the ^{87}Sr/^{86}Sr ratio of water tended to shift toward the ^{87}Sr/^{86}Sr ratio of rock.

The ^{87}Sr/^{86}Sr ratio for the groundwater inevitably shifts toward the mean value for the country rock, although groundwaters ascending rapidly in open channels are unlikely to reach complete equilibrium (Stettler and Allégre, 1978; Graham, 1992). Because water moves between the overlying and underlying aquifers, potential variation of the ^{87}Sr/^{86}Sr ratio exists, depending on the relative proportions of leakage from above and below

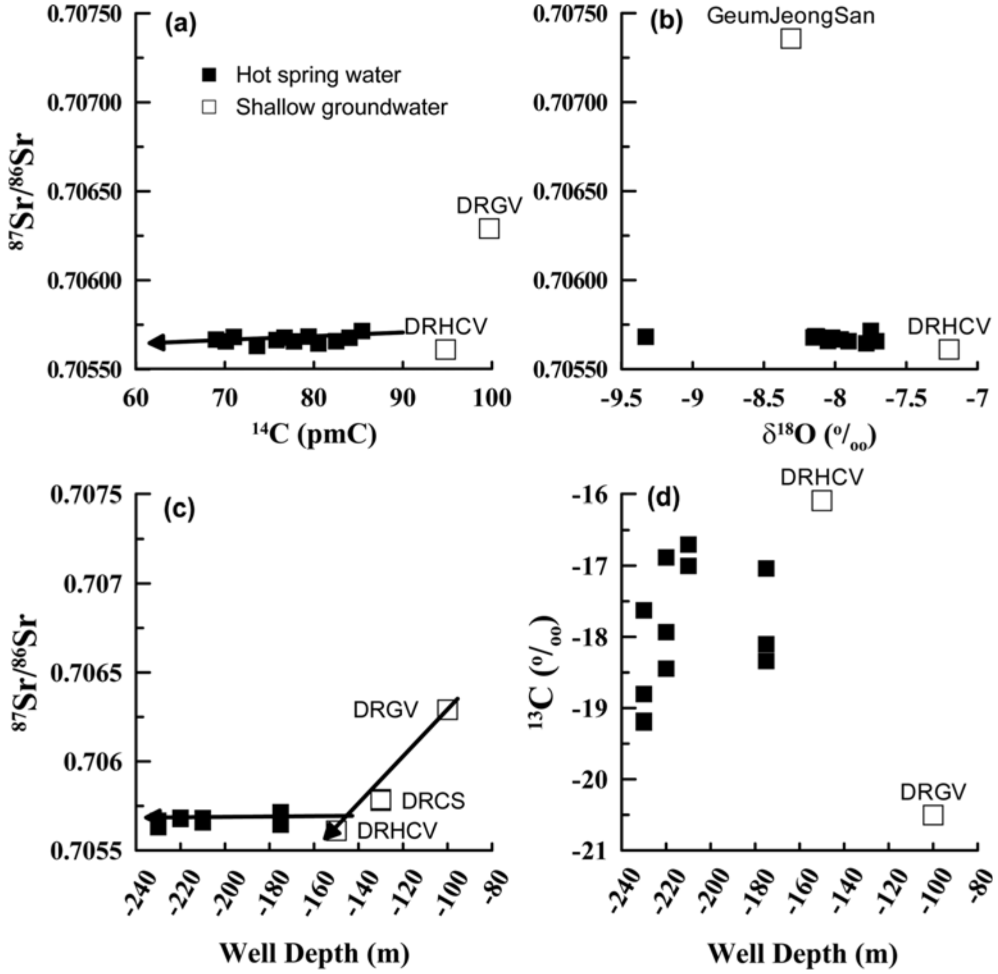

Fig. 6. Plots of the ^{87}Sr/^{86}Sr ratio vs. (a) ^{14}C, (b) δ^{18}O, and (c) the well depth, and (d) ^{13}C vs. the well depth for the Dongrae hot spring water.

(Woods et al., 2000). Franklyn et al. (1991) suggested that ^{87}Sr/^{86}Sr ratios in deep groundwater in felsic plutons are affected by interactions with plagioclase, and a low ^{87}Sr/^{86}Sr ratio in the groundwater reflected dissolution of very Sr-rich and Rb-poor plagioclase. Ishikawa et al. (2007) also suggested that ^{87}Sr/^{86}Sr ratios in groundwater may be affected by the mineral grains, because of the difference in the initial Rb/Sr ratio if the reacting rock is an old felsic rock.

^{87}Sr/^{86}Sr data for minerals in the granite near the Dongrae and Haeundae hot spring area are very few. Lee (1991) reported the Rb-Sr whole rock age of 83.9 Ma with an initial ^{87}Sr/^{86}Sr ratio of 0.70538 ± 0.00021 for the Kimhae granite. They also reported ^{87}Sr/^{86}Sr ratios of 0.70567 ± 0.00001(2σ) and 0.70563 ± 0.00001(2σ) for hornblende and plagioclase of the granite. The Kimhae granites occur in the western part of the Dongrae hot spring area. The age and initial ^{87}Sr/^{86}Sr ratio of the Kimhae granite are similar to those of the Dongrae hot spring-bearing granite reported by Yun et al. (2005). The ^{87}Sr/^{86}Sr ratios for the Dongrae hot spring waters were similar to those of the plagioclase and hornblende in the Kimhae granite. Therefore, the ^{87}Sr/^{86}Sr ratios in the Dongrae and Haeundae hot spring waters suggest that they may be affected mainly by plagioclase or hornblende in the old Cretaceous granite. Then, Lee et al. (2008) argued that the hot spring aquifer in the Dongrae area may be a nearly closed system with respect to the upper cold groundwater aquifer system. The steady values of ^{87}Sr/^{86}Sr ratio from the Dongrae and Haeundae hot spring waters (see Tables 1 and 2; Figs. 4a and 5a) support the argument that the hot spring waters were not affected by the upper cold groundwater aquifer system. Therefore, relatively constant ^{87}Sr/^{86}Sr ratio and a long residence age by the ^{14}C data of the hot spring waters might indicate that the Dongrae and Haeundae hot spring waters should be an old groundwater heated by a paleo-heat source rather than present heat source under the crust.

Generally, the hot spring waters are heated by geothermal heat like 1) the heat from the magma source, 2) the heat from the geothermal gradient, 3) the heat from the earth's mantle, 4) radioactive decay. Jeong et al. (2008, 2009) showed that, ^{3}He/^{4}He ratios from most of the hot springs in the granite area have characteristic of crustal source, and ^{40}Ar/^{36}Ar ratios of the hot springs are in the range of an atmo-

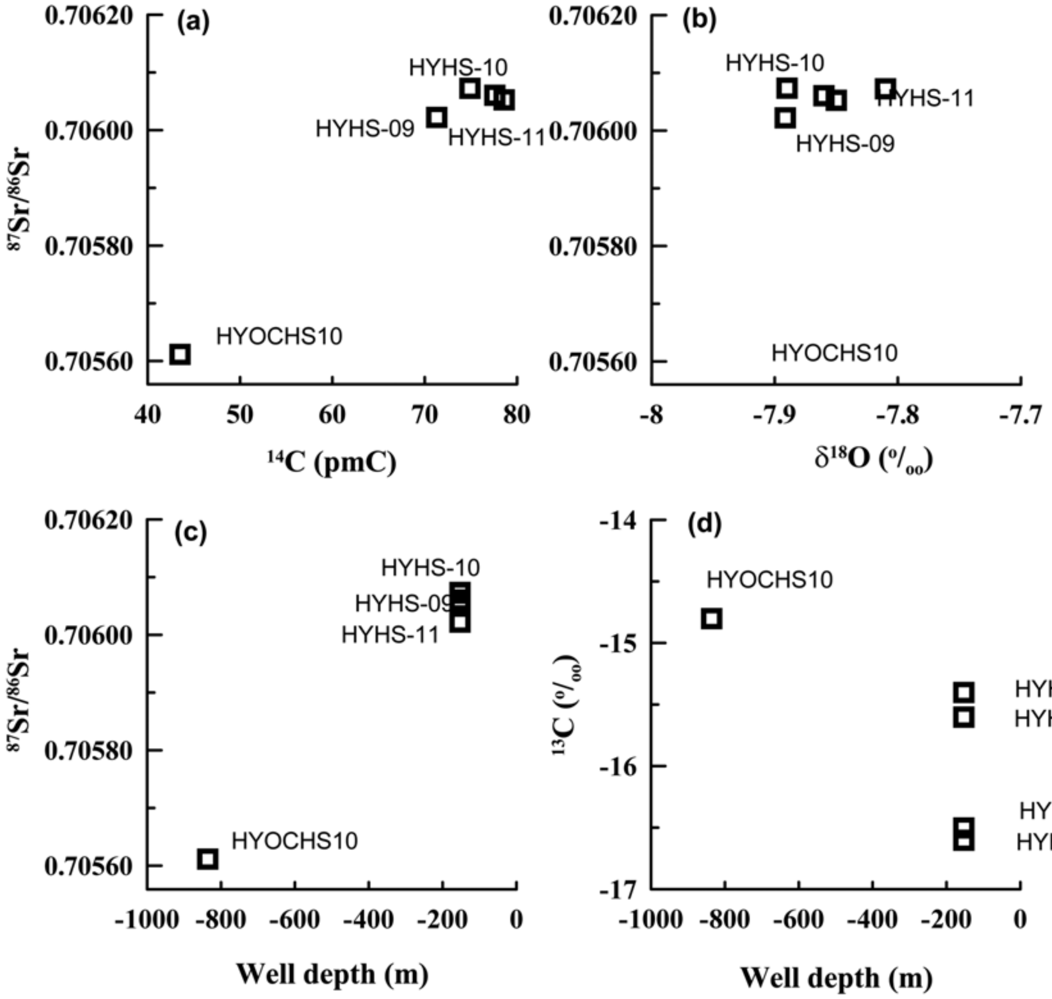

Fig. 7. Plots of the $^{87}Sr/^{86}Sr$ ratio vs. (a) ^{14}C, (b) $\delta^{18}O$, and (c) the well depth, and (d) ^{13}C vs. the well depth for the Haeundae hot spring water.

sphere source. This indicates that the heat source of the hot springs in the granite area in the southern part of Korean Peninsula might not have a relationship with the heat source from the mantle. And, there is no volcanic activity in the Korean Peninsula. Hence, at least, we can neglect two factors of the heat sources mentioned above. And it suggests that we can consider only other two factors, i.e., the heat from the geothermal gradient or the radioactive decay. At present, it may be difficult to precisely determine the heat source for the hot spring water in granitic areas, such as the Dongrae and Haeundae. It is true that there remain several questions regarding the heat source of the hot spring water in the granite area as here remain several questions to be addressed.

5.3. Geochemical Significance of the $^{87}Sr/^{86}Sr$ Ratio, ^{14}C, $\delta^{18}O$, and Well Depth

In the previous section, the oxygen and hydrogen isotope data, as well as geochemical characteristics, indicate that the hot spring water is likely derived from meteoric water that was affected by sea water. Furthermore, the ^{14}C data indicates that the residence time of the hot spring water in the aquifer is >1000 years.

If the $^{87}Sr/^{86}Sr$ ratio of the groundwater with depth varies, the ratio reflects interactions with the reactant rocks in the aquifer, because the $^{87}Sr/^{86}Sr$ ratio of the groundwater is fundamentally identical to that of the host rock equilibrated with the groundwater (Faure and Mensing, 2005). Then, Ishikawa et al. (2007) noted that discrepancies in the $^{87}Sr/^{86}Sr$ ratio of the groundwater in reactions between old rock and the groundwater might arise from differences in minerals solubility.

Figures 6 and 7 are correlation diagrams among the $^{87}Sr/^{86}Sr$ ratio, ^{14}C, $\delta^{18}O$, and well depth for the Dongrae and Haeundae hot spring waters. $^{87}Sr/^{86}Sr$ ratios for the Dongrae hot spring water did not vary with the ^{14}C concentration or well depth (Figs. 6a and c). However, those for the shallow groundwater decreased with ^{14}C concentration and well depth and also showed different trends than the $^{87}Sr/^{86}Sr$ ratio for the Dongrae hot spring water. In contrast, the $^{87}Sr/^{86}Sr$ ratio for the Haeundae hot spring water decreased with the ^{14}C concentration. The $^{87}Sr/^{86}Sr$ ratios for the Dongrae and Haeundae hot spring waters were lower than that of sea water (0.7092). Then, the difference of Mg concentration between hot spring waters and sea water geochemistry from the piper diagram (Fig. 2) indicates that the current sea water may not affect the hot spring waters from the Dongrae and Haeundae areas.

First of all, the relationships among the $^{87}Sr/^{86}Sr$ ratio, ^{14}C concentration, and well depth for the Dongrae hot spring water, at least, indicate that the Dongrae hot spring aquifer is not affected by shallow groundwater aquifer (Fig. 6). Due to lack of the cold groundwater data, it is difficult to mention the relationship between the Haeundae hot spring and shallow groundwater aquifers. However, Figures 7a and c seem to suggest that the sample HYHS should be slightly affected by shallow cold groundwater rather than sample HYOCHS due to its shallow depth.

These results collectively indicate that the hot spring aquifer in the Dongrae and Haeundae area may be a nearly closed system with regard to the upper cold groundwater aquifer system as well as sea-water. The results also suggest that $^{87}Sr/^{86}Sr$ ratios for deep groundwater such as the Dongrae and Haeundae hot spring waters would not vary for long time if there is no some break such as current sea-water intrusion or artificial injection of the shallow groundwater development. Therefore, it can be concluded that the sea-water composition of the hot spring water is due to mixing by paleo-sea-water intrusion or trapping, rather than current sea-water intrusion.

In this study, we could confirm that differing $^{87}Sr/^{86}Sr$ ratios for deep groundwater may be an indicator of the geochemical environments of different groundwater aquifers and the relative inputs from these aquifers. Our results showed that monitoring of the $^{87}Sr/^{86}Sr$ isotope ratios of the hot spring waters may be valuable in revealing the heat source of the high-temperature deep groundwater in the granitic area.

6. SUMMARY AND CONCLUSIONS

Over the last 11 years (2004–2014), we have analyzed the chemical composition, $\delta^{18}O$, δD, ^{13}C, ^{14}C, and $^{87}Sr/^{86}Sr$ ratios in waters from two hot springs (Dongrae and Haeundae) in the Busan area, Korea, to identify the heat source for the hot spring water in the Cretaceous granitic area. Geochemically, the Dongrae and Haeundae hot spring waters are Na-Cl type ranging with 1,095–2,100 μS/cm and 6,350–7,540 μS/cm, respectively. The oxygen and hydrogen isotopes in the hot spring waters indicate a meteoric origin. The $^{87}Sr/^{86}Sr$ ratio for the Dongrae and Haeundae hot spring waters have been nearly constant for 11 years, suggesting that the hot spring water may be nearly equilibrium with the aquifer.

Our results indicate that the $^{87}Sr/^{86}Sr$ ratio of the groundwater can be used as a tracer for clarifying the equilibrium state of water-rock interactions between cold shallow groundwater and high-temperature deep groundwater, and for determining the heat source of hot springs in granite areas.

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