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Rare Earth Elements and Sr-Nd-Pb Isotopic Analyses of the Arima Hot Spring Waters, Southwest Japan: Implications for Origin of the Arima-type Brine

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Abstract

Rare earth elements (REEs) and Sr-Nd-Pb isotopic compsotions of the Arima hot spring waters, a specific type of deep-seated brine (up to 6 wt.% NaCl) in the non-volcanic fore-arc region of southwest Japan, have been analyzed in order to discuss their source materials and origins. We have first examined the matrix effect associated with variable salinity (0 to 5 wt.% NaCl), and found that above 1 wt.% NaCl, the intensity of REE in ICP-MS measurement is drastically reduced due to the matrix effect. Accordingly, we have diluted the sample waters to contain ~0.6 wt.% NaCl, and then analyzed them by the standard addition method. The result shows that the abundance is appreciably high compared to near-surface waters, and exhibits almost a flat DMM-normalized pattern, which can be explained by mixing of a slab-derived fluid at relatively low temparature (400 to 500°C) and a near-surface water. The Sr-Nd-Pb isotopic compositions of the brineare consistent with the above interpretation on REEs, in that the deep brine is isotopically similar to a slab-derived fluid of the subducted Philippine Sea slab. These evidences suggest that the slab-derived fluid is upwelling even in the non-volcanic region possibly through a fault zone along the tectonic line.

Keywords: Brine; Hotspring; Slab-fluid; Arima; Subduction

Introduction

Research Article

Water cycling and fluid processes in subduction zones have been studied based on various observational, experimental and theoretical approaches. Stability fields of hydrous minerals in the subducting slabs, which have been constrained by the high-pressure experiments and thermodynamic estimates [1,2] are thought to primarily control the fluid supply at depth. Migration and re-distribution of the slabderived fluids (hereafter referred to as slab-fluid) has been predicted by numerical modeling [3-5]. Existence of such fluids has been examined by seismic studies along the subducting slabs and within the overlying mantle wedge [6,7], and by petrological studies of arc magmas that fingerprint the geochemical characteristics of slab-derived fluids [8,9]. Relatively well-constrained trace element behaviors upon slab dehydration [10-12], as well as heavy isotope tracers (such as Sr, Nd and Pb isotopic ratios) that are not fractionated through the processes of subduction and dehydration, have been utilized to successfully quantify the amount and composition of slab-fluids [13-15].

In addition to these constraints on the fluid processes in subduction zones from the deep side (i.e., dehydration of subducted slabs), several evidences have been accumulated from the surface side, including studies on fluid inclusions in volcanic phenocrysts and plutonmetamorphic rocks [16-18], and thermal waters and gases [19]. Within this context, the Arima-typebrine, which is a type of non-volcanic hot spring water with high chlorine content (~40000 ppm) and oxygenhydrogen isotopic ratios similar to magmatic/metamorphic thermal waters [20], has been argued to have originated from deep-seated brine [20-22]. The Arima-type brine may provide in valuable information concerning a slab-fluid in a fore-arc region where no magmatic information is available [23,24]. High ³He/⁴Heratios and large fault zones associated with the Arima-type brine suggest their deep origin, e.g., in the subducting slab and/or the mantle [25-27].

However, in order to discuss the origin of Arima-type brine, in particular its connection to the slab-fluid, better geochemical characterization is required, including Sr-Nd-Pb isotopic analyses which have not been done so far. This study aims mainly at developing an analytical method for the brine, including rare earth element abundances and ⁸⁷Sr/⁸⁶Sr, ¹⁴³Nd/¹⁴⁴Nd, ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb and²⁰⁸Pb/²⁰⁴Pb ratios. High salinity and low abundances of the target elements and isotopes (except for Sr) in the brine require specific analytical procedures to be developed. Based on the newly obtained data, we then briefly discuss the relationship between the slab-fluid and the Arima-type brine.

Geological Setting of the Studied Area

The Arima hot springs, a typical locality of the Arima-type brine, are located in the southwest Japan arc (Figure 1a), where two oceanic plates, the Pacific plate and the Philippine Sea plate, subduct beneath the area from the east with a velocity of 9 cm/year for the former and from the southeast with a velocity of 4 cm/year for the latter. The depth of the slab surface beneath the Arima area is ~400 m for the Pacific slab, whereas 50 to 80 km with a large uncertainty for the Philippine Sea slab [28,29]. In spite of the active subduction, a Quaternary volcano is not formed here because the Pacific slab is too deep and the Philippine Sea slab is too shallow (Figure 1a) to fulfill the physiochemical conditions for arc magma generation [30].

In southwest Japan, there are many faults striking in NW-SW or NW-SE slipping dextrally associated with the Median Tectonic Line [31]. The Arima-Takatsuki Tectonic Line (ATTL in Figures 1a and Figure 1b) is one of these strike-slip faults that have formed in the latemid-Miocene [31]. The average slip rate for the eastern range-front segment of the tectonic line is estimated to be 0.5-1.5 mm/year dextrally and 0.1-0.8 mm/year vertically during the late Quaternary period [32]. As shown in the geological map of this area (Figure 1b), the basement

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Figure 1: Tectonic and geologic maps of the Arima area. (a) the map showing distribution of the Quaternary volcanoes (red circles) and geometry of the subducting Pacific and Philippine Sea slabs, with Itoigawa-Shizuoka Tectonic Line (ISTL) and Median Tectonic Line (MTL). The pinkish contour lines indicate the depth of the upper surface of Pacific slab (50 to 300 km depth with 50 km interval), while the purplish contour lines indicate that of Philippine Sea slab (10 to 200 km depth with 10 km interval). The aseismic parts are shown by the dotted line. (b) The geologic map around the Arima area after Maruyama et al. [31].

around the Arima area is composed of late Cretaceous felsic volcanic rocks (rhyolite) of the Arima Group, granitic rocks (Rokko Granite), and late Eocene to early Oligocene non-marine sedimentary rocks with rhyolitic tuff layers of the Kobe Group [33]. The Arima Group directly covers the Rokko Granite in the south of ATTL and the sedimentary rhyolitic rocks in the north of ATTL in the Arima area [34].

The Arima-type brines occur typically in the Arima area, as well as the Osaka and Kii areas broadly in the Kinki region of southwest Japan. In the Arima area, the brines seem to upwell through ATTL and the subsidiary faults [35], with at least seven hot spring sources, i.e., Ariake, Gosha, Gokuraku, Kinsen, Tansan, Tenjin, and Uwanari. These hot spring waters exhibit a wide compositional range, which can be explained by mixing of two types of meteoric waters and a "deep brine" component [21,22]. The deep brine component is characterized by high solute concentrations (i.e., high Na, Cl, K, etc.), distinct from the meteoric waters. Based on the high salinity, we focus on the hot spring water from "Ginsuiso Kinsen" with ~40000 mg/L Cl from a 600 m depth pipe [23], which is composed primarily of the deep brine component, and analyze its composition for REEs and isotopic ratios as below.

Chemical Analysis

Sample description

The "Kinsen" water sample, together with other spring waters in

the Arima area, was collected in December 2010, directly from the well pipe before the water is pooled in an approximately $3\times3\times3$ m tankto be oxidized and colored 'gold (Kin-iro)' for commercial use. Our sample water is visibly colorless, implying that it is less oxidized and has a lower propensity to precipitate the solutes. Kusuda et al. [23] have analyzed "Ginsuiso Kinsen" sample (which is the same brine with "Kinsen", although the sampling date is different), 12 solute elements/ components and isotopic ratios of H, He, C, O: e.g., ~40000 mg/L Cl, ~20000 mg/L Na, 51.0 mg/L Li, $\delta^{18}O=5.0\%$, $\delta D=-34.1\%$, which is close to the estimated "deep brine" composition on the tritium-free basis [21-23].

Analytical method for rare earth element composition in high salinity brine

The high salinity and solute concentrations in the brine may disturb quantitative analyses of the rare earth elements due to the matrix effect. Moreover, in several water samples, there are some visible particles (or un dissolved materials). Accordingly, for determining the precise REEs composition, we have tested several preparation procedures as follows:

- (1) raw and filtrated brine samples were first dried up, respectively. The condensed materials were chemically digested in the same way as the dissolution of silicate rocks after Yokoyama et al., and then diluted with HNO₃ suitable for ICP-MS analysis.
- (2) raw and filtrated brine samples were centrifuged, and their supernatant liquids were pretreated straight for ICP-MS analysis; whereas, the precipitates were chemically digested in the same way as above, and then pretreated with HNO₃ for ICP-MS analysis.

The ICP-MS analysis was conducted at the Department of Earth and Planetary Sciences, Tokyo Institute of Technology. In order to quantify the matrix effect due to high salinity, we synthesized thirty standard solution samples with the NaCl concentration from 0.0 to 5.0 wt.% with 0.5% increment, and the REEs concentration of 0, 0.1, 1 ppb by adding the standard solution XSTC-1 (SPEX CertiPrep Co. Ltd.). All the combination of NaCl and REEs abundances have been analyzed by ICP-MS under the same tuning condition. The results show linear correlations between the REEs concentration and the signal intensity (in count per second (cps) unit) for each NaCl concentration from 0 to 5% (Figure 2). The results also show that the intensity drastically decreases



Figure 2: Matrix effect of brine in ICP-MS measurement. Intensity (count per second) of ICP-MS measurement of La in synthetic standard solutions (0, 0.1 and 10 ppm) as a function of NaCl content (0.0 - 5.0% with 0.5% interval). The inset shows a magnified view near the origin.



Figure 3: Summary of pretreatment method for isotope analysis. (a) a brine sample added with concentrated original HNO₃ solution to dissolve all ingredients. (b) minor visible and dispersed precipitation occur after adding ammonia solution at pH 7-8. (c) the dried-up sample of the precipitate of (b). (d) the final solution.

when the NaCl concentration exceeds 1% (Figure 2). Accordingly, we have diluted the unknown samples of 0.2 mL with 1.7 mL pure water, and have applied the standard addition method with addition of XSTC-1 (0.1 ml) by 0, 0.1, 1 and 10 ppb to the diluted sample. As a result, the final diluted sample contained ~0.6 wt% NaCl.

In Figure 2, even for a specific NaCl content, a slight (less than ~5%) deviation from the perfect linear relation is recognized, indicating a complex matrix behavior that may also depend on the concentration of the target element. We therefore have performed ICP-MS analysis with the standard addition method using three combinations of XSTC-1 concentrations ((I) 0-1 ppb, (II) 0.1-1 ppb and (III) 0.1-10 ppb). As will be shown, the results show good reproducibility, confirming a weak nonlinear matrix effect when NaCl content is relatively low. It should be noted that Eu was not quantitatively measured due to significant interference from Ba oxide, which unfortunately prevents us from discussing the Eu anomaly.

Heavy isotopic composition in high salinity brine

We have analyzed the isotopic compositions of Sr, Nd and Pb (⁸⁷Sr/⁸⁶Sr, ¹⁴³Nd/¹⁴⁴Nd, ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb and²⁰⁸Pb/²⁰⁴Pb ratios) for the "Kinsen" brine. The target elements, i.e., Sr, Nd and Pb, have been extracted and separated from the brine sample by a co-precipitation method with iron (III) hydroxide (Figure 3). First, a sufficient quantity of highly purified and concentrated HNO₃ is added to the sample to dissolve, any existing precipitates (Figure 3a). Then, highly concentrated aqueous ammonia is added to neutralize the sample. When pH is between 7 and 8, some visible but dispersed precipitates occur, after 24

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hours which form a deposit that contains rare earth elements (Figures 3b and 3c). The recovery percentage of this method is more than 95% [36]. We have successfully obtained approximately 0.3 g precipitate from 500 ml brine. We dissolve the precipitate with HNO_3 (7M) (Figure 3d) [37] from which Sr, REEs and Pbare extracted by column separation (Figure 4). The extracted Sr, Nd and Pb werefinally analyzed by MC-ICP-MS installed at Earthquake Research Institute, The University of Tokyo.

Results and Discussions

REEs pattern of Arima brine

The results are listed in Table 1 and plotted in Figure 5a as compositional patterns normalized by depleted MORB mantle (DMM). The REEs abundances of the "Kinsen" brine (three lines with bluish

colors) are approximately three times lower than those of DMM, and show a flat pattern with a slight increase to both the left for light REEs and the right for heavy REEs. The results are fairly consistent with those from the previous study, shown as a black line in Figure 5a [38], in terms of the overall level and a slight increase towards the left for light REEs, although the previous result shows a slight decrease from middle to heavy REEs.

We now compare the results with the REEs abundances of possible "deep brine", in particular the slab-fluids, and examine whether the slab-fluids could be a source of deep brine. Figures 5b and 5c show estimated REEs compositions of aqueous fluids dehydrated from the major constituent materials of a subducting slab: i.e., altered oceanic basaltic crust (AOC) and sediment. In Figure 5b, the REEs abundances



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La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
0.9294	0.9744	0.1157	0.5044	0.3272	-	0.5618	0.087	0.6556	0.1491	0.5306	0.0753	0.6393	0.1154
0.8872	0.986	0.1379	0.4779	0.3224	-	0.5676	0.1206	0.6779	0.1809	0.4995	0.0996	0.5758	0.1435
0.7629	0.8493	0.0915	0.4096	0.2542	-	0.456	0.0774	0.566	0.1301	0.4093	0.0554	0.4725	0.0953
La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
151.36	414.95	61.435	242.5	38.134	8.4499	22.231	1.8234	20.621	3.8322	5.339	-	7.1975	0.58
232.71	572.69	74.414	264.01	38.134	8.2564	20.817	1.3685	18.401	2.7819	4.405	-	5.9307	0.48
230.08	362.17	85.083	323.08	44.178	10.018	48.357	5.8506	43.223	2.8905	16.724	1.1356	13.7	2.0647
62.399	120.05	-	295.5	6.908	1.4089	6.1252	0.87162	4.5372	-	1.8066	-	3.4	0.5368
La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
0.0061	0.0106	-	0.0258	0.0093	0.0039	0.0138	-	0.0849	-	0.0879	-	0.0911	0.0251
0.0444	0.0746	-	0.1396	0.0438	0.0171	0.057	-	0.2477	-	0.2198	-	0.2173	0.0548
0.3905	0.6343	-	0.8844	0.2398	0.0858	0.269	-	0.7991	-	0.5996	-	0.5625	0.129
3.6742	5.7647	-	5.9399	1.3845	0.4521	1.3336	-	2.6747	-	1.688	-	1.5002	0.3119
59.511	89.436	-	63.279	12.221	3.5623	9.7439	-	11.997	-	6.1057	-	5.074	0.9335
0.0001	0.0001	-	0.0006	0	0	0	-	0.0001	-	0.0001	-	0.0001	0
0.0005	0.0005	-	0.0033	0.0001	0	0.0001	-	0.0002	-	0.0001	-	0.0002	0.0001
0.004	0.0046	-	0.0211	0.0004	0.0001	0.0003	-	0.0007	-	0.0003	-	0.0006	0.0001
0.0373	0.0414	-	0.1416	0.0025	0.0005	0.0017	-	0.0022	-	0.001	-	0.0017	0.0003
0.6038	0.6429	-	1.508	0.022	0.0039	0.0123	-	0.01	-	0.0035	-	0.0056	0.001
La	Се	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
0.87	1.3	0.16	0.7	0.25	0.04	0.51	0.11	0.91	0.21	0.66	0.092	0.54	0.078
-	0.204	-	0.392	0.098	0.0045	0.108	-	0.0907	-	0.0458	-	0.039	0.006
-	0.0688	-	0.177	0.0437	0.0024	0.0607	-	0.0631	-	0.0356	-	0.0267	0.0037
0.024	0.0062	-	0.0373	0.0101	0.0016	0.0179	-	0.0275	-	0.0345	-	0.0511	0.0124
0.0407	0.0202	-	0.0346	0.00638	0.00068	0.00612	-	0.00606	-	0.00315	-	0.00309	0.00051
0.205	-	-	0.189	0.0364	0.0038	0.0382	-	0.0325	-	0.0198	-	0.0131	0.0018
0.405	0.0228	-	0.0326	0.00635	0.00069	0.00652	-	0.00698	-	0.00662	-	0.0108	0.0019
0.0282	0.00791	-	0.0351	0.0074	0.0011	0.0121	-	0.0152	-	0.0204	-	0.0351	0.0082
0.0445	0.0127	-	0.034	0.00698	0.0009	0.00718	-	0.00621	-	0.00452	-	0	0.00066
0.049	0.00388	-	0.0325	0.0054	0.00083	0.00574	-	0.00524	-	0.00422	-	0.00398	0.00071
0.316	0.951	-	0.466	0.081	0.0096	0.0959	-	0.0827	-	0.0447	-	0.042	0.00634
0.513	0.171	-	0.54	0.121	0.0101	0.115	-	0.0979	-	0.0737	-	0.0975	0.0157
0.192	0.55	0.107	0.581	0.239	0.096	0.358	0.07	0.505	0.115	0.348	0.06	0.365	0.058
La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
4.9804	6.042	-	18.1577	0.353	-	0.288	-	0.579	-	0.3702	-	-	0.0839
-	0.13	-	0.691	0.021	-	0.024	-	0.09	-	0.079	-	0.0937	0.0236
-	0.122	-	0.6298	0.024	-	0.029	-	0.087	-	0.0739	-	0.0857	0.0213
	La 0.9294 0.8872 10.7629 232.71 230.08 62.399 62.399 10.0061 0.0044 0.0001 0.0444 0.3905 3.6742 59.511 0.0445 0.0041 0.0005 0.0045 0.0045 0.0282 0.0405 0.205 0.0282 0.0405 0.0282 0.0405 0.0282 0.0405 0.0282 0.0405 0.0282 0.0405 0.0282 0.0405 0.0282 0.0405 0.0282 0.028	LaCe0.92940.97440.88720.9860.76290.8493LaCe151.36414.95232.71572.69230.08362.1762.399120.05LaCe0.00410.01060.4440.07460.39050.63433.67425.764759.51189.4360.00410.00010.00420.00010.00430.004140.00430.004140.00440.004210.03730.04140.60380.64290.4450.02420.0240.006210.04070.020210.04080.012170.04090.03160.19210.19210.1920.5511.30.17110.1920.5511.30.17110.1920.5511.4Ce1.5130.17210.4450.02140.5130.1710.1920.5511.4Ce1.5446.04221.551.51.50.1321.50.1321.50.1321.50.1321.50.1221.50.122	La Ce Pr 0.9294 0.9744 0.1157 0.8872 0.9860 0.1379 0.7629 0.8493 0.0915 La Ce Pr 151.36 414.95 61.435 232.71 572.69 74.414 230.08 362.17 85.083 62.399 120.05 6.1 14 0.0746 6.1 0.0061 0.0106 - 0.044 0.0746 6.1 0.044 0.0746 6.1 0.041 0.0106 - 0.041 0.0106 - 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Table 1: Rare earth element abundances of the Arima hot spring water "Kinsen" brine are listed in the "Analyzed abundance" row. Analytical precisions for all elements are below 2% (2SD). The compositions of AOC-derived fluids and sediment-derived fluids are calculated forwardly starting from the compositions of raw materials before subduction for Pacific and Philippine Sea plates, respectively, as shown in the "Calculated abundance" row [39]. The estimated compositions of fluids derived from subducting slab at low temperatures for Philippine Sea plate, are shown in the "Estimated abundance" row. The compiled data used in this study are also listed in the "Compiled data" row. The optimized data to the analyzed abundance by a mixing among slab-derived fluid and river water are listed in the "Mixed fluid" row.

of AOC-derived fluids ("PHS" derived from the Philippine Sea slab and "PAC" from the Pacific slab) beneath volcanic arc exhibit high concentrations [39], whereas those dehydrated at lower temperatures beneath the fore-arc region exhibit significantly lower abundances due to the large temperature dependence of partition coefficient between fluid and residual solid [12,15,40]. derived fluids beneath the volcanic region exhibit high concentrations (Figure 5c) [39]. However, unlike the AOC system, the temperature dependence of partition coefficients between fluid and sediment is not well constrained. Therefore, we assume that, with a temperature decrease, the same degree of REEs concentration reduction occurs in the sediment system (Figure 5c) as in the basaltic system (Figure 5b). This is a crude assumption, and these results (shown by dotted lines in

Likewise, the estimated REEs compositions of the sediment-

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Figure 5c) should be regarded as preliminary estimates.

In any case, deep brine must mix with the near-surface water, as evidenced by δ^{18} O- δ D- tritium systematics [20-22]. In Figure 6, we estimate a best fit composition of fluid that mixes a slab-fluid (that consists of a mixture of AOC-derived fluid (Figure 5b) and sediment-derived fluid (Figure 5c) and the river waters in the Rokko Mountains (Figure 5d) [41]. The preliminary result suggests that the REEs abundances of the "Kinsen" sample (Figure 5a) may be broadly explained by the mixing of approximately 81-92% fluid derived from the Philippine Sea slab (at 400°C) or 56% at 510°C (Figure 6), with the addition of river water, being consistent with the previous studies based on the δ^{18} O- δ D- tritium systematics, although the misfit is rather large at this stage.

Heavy isotopic composition of Arima brine considered in arc systems

The results of Sr-Nd-Pb isotopic analyses are listed in Table 2. In addition to the "Kinsen" brine, "Tenjin" and "Tansan" have been also

analyzed for examining heterogeneity among the hot spring sources in the Arima area. In order to test the slab-origin hypothesis, the isotopic compositions are compared with the isotopic compositions of the slabfluids (PHS-fluid and PAC-fluid), as well as the subducted materials of AOC and sediment (Figure 7). The "Kinsen" data are plotted in the range of slab-fluid derived from the Philippine Sea slab (PHS-fluid), being consistent with the REEs arguments in the last section. It is noted that the PHS-fluid is mainly derived from sediments [9] and therefore resembles the sediment in isotopic composition.

While the ²⁰⁷Pb/²⁰⁴Pb vs. ¹⁴³Nd/¹⁴⁴Nd diagram is sufficiently sensitive to resolve the type of slab-fluids (Figure 7), the Sr and Nd isotopic diagram is useful to discuss the contribution from the crustal rocks, especially the granitoids that form the basement of the Arima area. Figure 8 shows the Sr-Nd isotopic compositions of the basement granitoids in the southwest Japan arc [42,43], as well as those of slab-fluids (PHS-fluid and PAC-fluid) and the subducted AOC and sediment. The "Kinsen" data appear to be plotted near the PHS-fluid composition.

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Figure 6: DMM (depleted MORB mantle)-normalized REE compositions of calculated mixed fluid between slab-fluid derived from the Philippine Sea slab and the river water in Figure 5.

Sample name	87Sr/86Sr	¹⁴³ Nd/ ¹⁴⁴ Nd	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb
Tenjin (1-4)	0.708516	-	18.2705	15.5714	38.4193
Tansan (3-1)	0.708916	0.512318	18.5874	15.6295	38.8785
Kinsen (4-3b)	0.708958	0.5123	18.4751	15.6307	38.705
Kinsen (4-3c)	0.708988	0.512333	18.4857	15.6362	38.7288
± 2 s.e.	0.00008	0.000008	0.002	0.003	0.004

Table 2: Sr-Nd-Pb isotopic compositions of the Arima hot spring waters from the "Kinsen", "Tenjin" and "Tansan" hot spring sources. The Nd isotopic ratio of "Tenjin" was not obtained. Other data are compiled from Nakamura et al. [14].



Figure 7: Pb-Nd isotopic composition of the Arima hot spring waters (red crosses) compared with the volcanic rocks in central Japan of Nakamura et al., [14]. Green and orange bands with black line represent the mixing of slabderived fluid with the mantle: green is for the Pacific slab (PAC), and orange for the Philippine Sea slab (PHS). The ellipsoid represents the compositional range for each end-component. Small circles represent the Quaternary volcanic rocks in central Japan for comparison, which can be explained by the mixing between slab-derived fluids (PAC- and PHS-fluids) and the mantle (DMM).



At the same time, the data are included in the broad compositional range of the basement granitoids, although the basement granitoids in areas adjacent to Arima (box coded in dark purple in Figure 8) deviate from the "Kinsen" data, and we may not identify the unique source material of "Kinsen" in this case. However, considering the high ³He/⁴He ratio of the Arima brine [27], it is likely that the Arima brine is related to the PHS-fluid deep in the mantle and the subducted slab.

Conclusion

The new analytical procedures for REEs and Sr-Nd-Pb isotopic compositions have been presented in this study, based on which typical Arima-type brines have been analyzed. The most plausible interpretation for both the REEs abundance and the isotopic ratios is that the deep brine has been derived from a subducted Philippine Sea slab, being consistent with the oxygen, hydrogen and helium isotopic compositions. This may indicate that slab-fluids may ascend in the fore-arc region, in addition to the volcanic region where the slab-fluid triggers arc magmatism, possibly through a large fault zone along the tectonic line. Several Arima-type brines with high ³He/⁴He ratios in other areas along the Median Tectonic Line in southwest Japan (e.g., Kazahaya et al. this special issue) may also suggest a similar origin, which provides invaluable information on slab-fluid in non-volcanic regions.

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