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Key Points:

- The stable strontium isotopes for the Arima-type saline water show light isotope enrichment and are different from those of local rocks
- The strontium isotopes of the saline water reflect the primary characteristics of slab-fluids without an isotopic overprint in the crust
- The light strontium isotope-enriched signature of the saline water suggests isotope fractionation associated with slab-fluid generation

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Strontium Isotope Characteristics ($\delta^{88/86}\text{Sr}$, $^{87}\text{Sr}/^{86}\text{Sr}$) of Arima-Type Brines Originated From Slab-Fluids

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Abstract In the southwest Japan forearc, slab-fluids produced from subducted materials migrate to crustal levels and appear as deep-seated brine. We have analyzed for the first-time stable strontium isotopes in non-volcanic spring water with high salinity, referred to as Arima-type saline water that likely originate from slab-fluid that upwelled along major faults. The stable strontium isotope compositions of the saline water are isotopically light ($\delta^{88/86}\text{Sr} = 0.122\text{--}0.157\text{‰}$) and different from those of local bedrock and near-surface water. The light strontium-enriched and radiogenic signature of the saline water reflects the primary characteristic of slab-fluids without an isotopic overprint in the crust. The Arima-type brines show signatures of slab-fluids at forearc depth, which is different from the slab-fluids at subarc depth estimated from arc lavas. The characteristic features of the Arima-type brines are explained by a larger contribution of subducted sediments and strontium isotope fractionation during fluids generation at shallower depth.

Plain Language Summary Slab-fluids are likely sourced from oceanic crust and sediments, such as carbonate, chert, and clay, materials that are subducting into the mantle at convergent boundaries, which is a process that triggers arc volcanism and transports elements/water. The fluids released in the deep subduction zone appear to migrate to the Earth's surface and upwell along major faults in western Japan as spring water with high salinity, referred to as Arima-type saline water. The stable strontium isotope compositions of the Arima-type saline water are different from those of surrounding rocks, interpreted as reflecting the isotopically light signature as geochemical characteristics of slab-fluid. The lighter strontium enrichments are thought to be the result of involvement of subducted crust and sediments, and likely reflect the mass dependent isotope fractionation with a preferential extraction of the lighter isotopes during slab-fluid generation.

1. Introduction

Slab-fluids, which are generated by the dehydration of subducted oceanic crusts and overlying sediments, play an indispensable role in element mobility, mass transfer, and water (H_2O) budgets in the subduction zone. Nonvolcanic hot spring water with high salinity upwells along major faults in the forearc, which has no Quaternary volcanism in western Japan. This water, referred to as the “Arima-type saline water,” has magmatic oxygen, hydrogen, and carbon isotopic compositions (e.g., Masuda et al., 1986; Matsubaya et al., 1973), suggesting that it possibly originated from slab-fluids (e.g., Kazahaya et al., 2014; Kusuda et al., 2014; Kusuhara et al., 2020; Morikawa et al., 2016; Nakamura et al., 2014).

One of the fluid-mobile elements, strontium is a particularly useful tracer of mass transfer in subduction zones as the presence of both radiogenic and mass-dependent isotope systems enables different processes to be resolved. The strontium isotopic data of the Arima saline water ($^{87}\text{Sr}/^{86}\text{Sr} = 0.70853\text{--}0.70879$; Kusuda et al., 2014) cannot be determined to be representative of slab-fluids because the $^{87}\text{Sr}/^{86}\text{Sr}$ values overlap not only with the slab-fluids estimated as a mixed component between the subducted sediments ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7100$; Plank et al., 2007) and the altered oceanic crust (AOC) ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7035$; Hickey-Vargas, 1991; Chen et al., 2021) but also with those of the basement, the Rokko granite ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7084\text{--}0.7440$; Terakado & Nohda, 1993). Because stable strontium isotope values ($\delta^{88/86}\text{Sr} = [(^{88}\text{Sr}/^{86}\text{Sr})_{\text{sample}} / (^{88}\text{Sr}/^{86}\text{Sr})_{\text{SRM987}} - 1] \times 10^3$) reflect isotopic fractionation without time-integrated effects, they provide a different perspective of the resultant radiogenic strontium ($^{87}\text{Sr}/^{86}\text{Sr}$)

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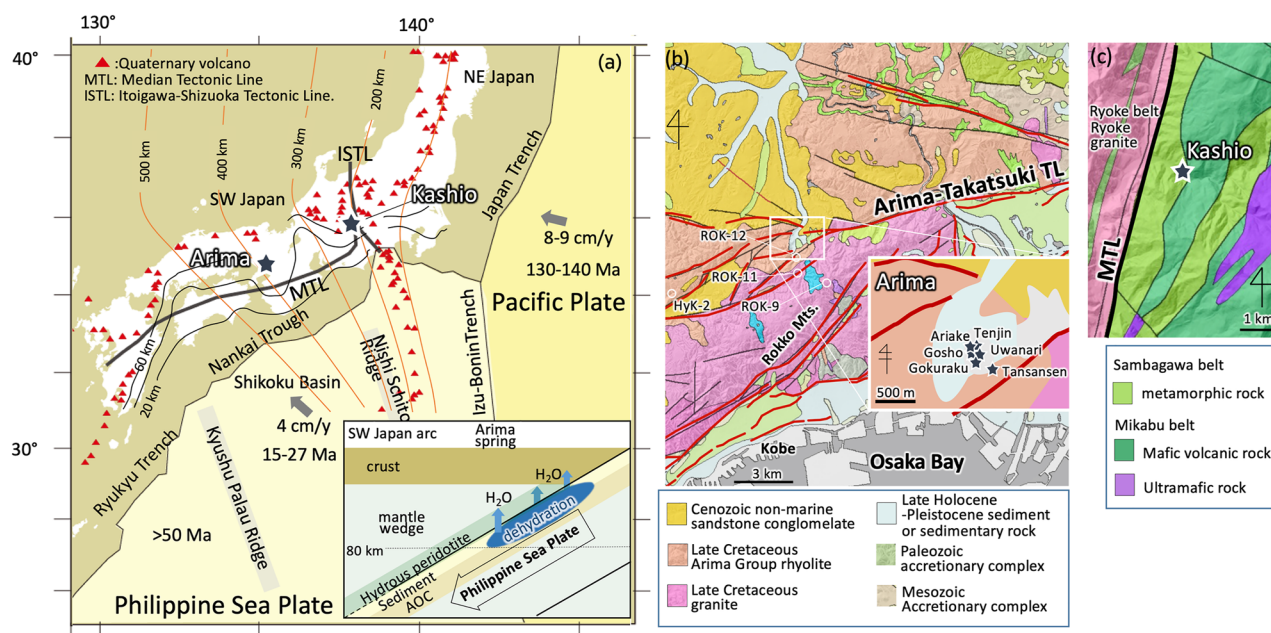


Figure 1. (a) Tectonic setting in Japan and location of the study area modified after Nakamura et al. (2019). The PHS plate is subducting beneath the Arima and Kashio areas. The subducted older cold Pacific plate exists under the PHS plate. The black and red contour lines indicate the depth of the upper surface of the subducted PHS plate and the subducted PAC plate, respectively (Hirose et al., 2008; Nakajima & Hasegawa, 2007). The schematic diagram shows aqueous fluid transportation from the subducted plate to the Arima area (modified from Kazahaya et al. (2014) and Nakamura et al. (2019)). The fluid is generated from the subducted sediment and the AOC. Approximate depth ranges of fluid dehydration are after Nakamura et al. (2019). (b) Geologic map around the Arima area with active faults (the Research Group for Active Faults of Research Group for Active Fault of Japan, 1991; Maruyama & Lin, 2002). The star marks the location of the Arima springs. The circles indicate the location of rock samples (c) Geologic map of the Kashio area with the MTL. The star marks the location of the Kashio springs. Geologic maps are from the Geological Survey of Japan, AIST (2015).

composition. The dual isotope systems, that is, $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{88/86}\text{Sr}$, can assess isotopic disturbances at crustal level and clarify geochemical characteristics of the Arima-type saline water as a fingerprint of slab-fluids.

This study presents the first application of stable strontium isotopes for the Arima-type saline water. The saline water samples were collected from two study areas, the Arima hot springs in Hyogo Prefecture and the Kashio mineral springs in Nagano Prefecture, Japan. Stable and radiogenic strontium isotope compositions of bedrock and surface water around the Arima area were also analyzed to clarify whether their isotope signatures reflect those from slab-fluids or a result of chemical evolution during migration to the surface. Based on results from the dual strontium isotope systems, we evaluate the contribution of subducted materials to slab-fluids and discuss the fluid-mobile strontium behavior during fluid generation within the forearc environment.

2. Samples and Geological Background

The Arima-type springs are distributed along the Median Tectonic Line (MTL), and subsidiary faults that form along the southwest Japan arc, which is characterized by the absence of Quaternary volcanoes despite the active subduction zone. The locations of the studied springs, the Arima hot springs and the Kashio mineral springs, are shown in Figure 1. In the subduction system, depths of the Philippine Sea (PHS) slab are estimated to be 50–70 km with large uncertainties beneath both areas; the old Pacific (PAC) slab surface depth of ~400 km in the Arima area and 200–300 km in the Kashio area (Hirose et al., 2008; Nakajima & Hasegawa, 2007).

The basement rocks around the Arima springs are mostly composed of the Late Cretaceous Rokko granite and the rhyolitic rocks of the Arima Group (Arai, 2007). The springs upwell through the Arima-Takatsuki Tectonic Line. Five sample sites are distributed over a narrow area of less than 0.25 km² (Figure 1b); the Uwanari, Goshō, and Tenjin springs were sampled in 2008 and the Tenjin, Gokuraku, and Ariake springs were sampled in 2018. Carbonate samples (calcite and aragonite) precipitated at high temperature (~90°C) were collected from thick-layer deposits on the outer surface of spring water tanks and water-pumping pipes. A cold spring water sample with low salinity (Na = 18 mg/L) was collected from the Tansansen spring which upwelled from

a shallower meteoric aquifer. The chemical and isotopic characteristics of these springs are presented in Kusuda et al. (2014). Descriptions of the analyzed Rokko granite and rhyolitic welded tuff samples appear in Terakado and Nohda (1993).

The basement of the Kashio area mostly comprises the Ryoke granites, Sanbagawa metamorphic rocks, and Mikabu mafic rocks. Springs upwell through the main boundary fault of the MTL (Ishikawa et al., 2014; Kawamoto et al., 2013). The saline water samples were collected from the springs at two sites (Kashio A and Kashio B; Figure 1c). The chemical and isotopic characteristics of these springs are presented in Kusuha et al. (2020).

3. Methods

3.1. Sample Preparation

Sample preparation and chemical separation of strontium mostly followed the procedures from Neymark et al. (2014). Water samples were acidified by adding concentrated nitric acid. Carbonate samples were dissolved using dilute hydrochloric acid. Silicate samples were digested using a mixture of hydrofluoric acid and nitric acid. Samples were re-dissolved in 6 M HCl from which aliquots were taken for spiked and unspiked analyses. The former was mixed with the double spike (DS) prior to chromatography. The silicate sample solution was first passed through a 500- μ L anion exchange column (BioRad AG1-X8, particle size: 37–74 μ m) to remove Fe (III). The sample solution was passed twice through a column filled with 150 μ L of Eichrom Sr resin (particle size: 50–100 μ m) on top of 350 μ L of polystyrene divinylbenzene adsorbent (Mitsubishi MCI GEL CHP-20P, particle size: 37–74 μ m). The chemical yield of strontium during extraction chromatography was sufficiently high (>95%). Total procedural blanks for strontium were $\sim$ 60 pg and considered to be negligible for most of the samples in this study.

3.2. Strontium Isotope Analyses

Measurement methods and ^{84}Sr - ^{87}Sr DS preparation followed those of Neymark et al. (2014). Two spikes enriched in ^{84}Sr (from Oak Ridge National Laboratory) and in ^{87}Sr (from ISOFLEX, USA) were used. The DS calibration was repeatedly conducted using the NIST SRM987 + DS mixture analyses and unspiked SRM987 analyses. Isotope abundance of our double spike was calculated using the DELTA_SR program developed by Nik Meľnikov (Neymark et al., 2014). The isotopic abundances of our DS are $51.56456 \pm 0.0021\%$ ^{84}Sr , $0.54916 \pm 0.0007\%$ ^{86}Sr , $43.725098 \pm 0.0022\%$ ^{87}Sr , $4.16118 \pm 0.0036\%$ ^{88}Sr (uncertainties = 2 standard deviations [2s]).

Strontium isotopic measurements were made in static mode, using multi-collector thermal-ionization mass spectrometers (TIMS) at Kumamoto University and at the National Museum of Nature and Science both equipped with $10^{11} \Omega$ registers for the Faraday cups. Approximately 500 ng of strontium was loaded on outgassed tungsten filaments with a tantalum activator. Analyses were carried out using an ^{88}Sr beam intensity of 8×10^{-11} A for 10 cycles of 15 block ratio measurements with a 16.8-s integration time and a 3-s idle time. Before each block, the baseline (deflected beam) was measured for 80 s. Unspiked strontium isotope analyses were normalized to $^{88}\text{Sr}/^{86}\text{Sr} = 8.375209$. The final $^{87}\text{Sr}/^{86}\text{Sr}$ results for the samples were adjusted to an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.710250 for the National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) 987.

Using the DELTA_SR program (Neymark et al., 2014), the stable strontium isotope compositions were calculated and compared to the $(^{88}\text{Sr}/^{86}\text{Sr})_{\text{SRM987}}$ value of 8.375209 via $\delta^{88/86}\text{Sr} = [(^{88}\text{Sr}/^{86}\text{Sr})_{\text{sample}} / (^{88}\text{Sr}/^{86}\text{Sr})_{\text{SRM987}} - 1] \times 10^3$. Repeated analyses of multiple prepared (sample + DS) mixtures have been performed. Each $\delta^{88/86}\text{Sr}$ result represents a value calculated for each (sample + DS) mixture. Weighted averages for replicate DS-spiked analyses that used the within-run standard deviation (s) as a weighting factor were calculated with the *Isoplot/Ex* program (Ludwig, 2009). During the course of this study, replicate analyses of multiple seawater (NASS-6)-DS mixtures yielded an intermediate precision for the measurement results of $\delta^{88/86}\text{Sr} \sim 0.03\%$ (2s, $n = 7$). The weighted average $\delta^{88/86}\text{Sr}$ value for NIST SRM987 was $0.008 \pm 0.010\%$ (2s, $n = 42$). Based on the $\delta^{88/86}\text{Sr}$ values of NIST SRM987 measured at the same session, all $\delta^{88/86}\text{Sr}$ results for unknown samples were bias corrected (assuming $\delta^{88/86}\text{Sr}_{\text{SRM987}} \equiv 0$). The “true” $^{87}\text{Sr}/^{86}\text{Sr}^*$ ratio refers to the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio measured and normalized to the ^{84}Sr - ^{87}Sr DS. This ratio was not normalized to an $^{88}\text{Sr}/^{86}\text{Sr}$ ratio of 8.375209 (see Krabbenhöft et al., 2009).

4. Results

The strontium isotopic results are presented in Table 1. The $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{88/86}\text{Sr}$ results for reference materials are consistent with the previous DS-TIMS measurements (Krabbenhöft et al., 2009; Neymark et al., 2014).

Table 1
Strontium Isotopic Results for Spring Water, Carbonate Precipitates, Seawater, and Rock Samples

Sample ID	Weight [g]	Sr [μg/g]	⁸⁷ Sr/ ⁸⁶ Sr		⁸⁷ Sr/ ⁸⁶ Sr*		δ ⁸⁸ / ⁸⁶ Sr [‰]		
Spring water in Arima area									
Uwanari (KNKW_08_002)	60.54	33.3	0.708536	±5 ^a	0.708590	±41 ^b	0.153	±42 ^b	(n = 3)
Gosho (KNKW_08_003)	59.27	15.4	0.708529	±2	0.708577	±54	0.128	±51	(n = 2)
Tenjin '08 (KNKW_08_004)	60.31	36.1	0.708532	±2	0.708575	±49	0.122	±49	(n = 2)
Tenjin '18 (KNK_18_B206)	60.04	37.7	0.708536	±5	0.708590	±44	0.146	±43	(n = 3)
Gokuraku (KNK_18_B207)	59.40	17.0	0.708535	±5	0.708587	±40	0.140	±43	(n = 3)
Ariake no. 1 (KNK_18_B205)	59.24	18.9	0.708527	±5	0.708581	±51	0.145	±53	(n = 2)
Tansansen (KNKW_08_001)	176.33	0.142	0.708436	±5	0.708543	±25	0.248	±48	(n = 2)
Spring water in Kashio area									
KashioA (KNK_16_M030)	50.46	162	0.709204	±5	0.709249	±47	0.149	±52	(n = 2)
KashioB (KNK_16_M029)	50.60	82.3	0.709201	±4	0.709256	±39	0.157	±41	(n = 3)
Modern seawater									
NASS-6 (CNRC)			0.709177	±5	0.709327	±33	0.393	±26	(n = 7)
Rocks	[mg]								
Rokko Granite (ROK-9-WR)	91.57	204	0.708516	±5	0.708607	±42	0.248	±45	(n = 3)
Rokko Granite (ROK-11-WR)	135.62	231	0.708413	±3	0.708488	±23	0.246	±47	(n = 2)
Rhyolitic welded tuff (ROK-12-WR)	171.24	138	0.711293	±5	0.711381	±42	0.243	±42	(n = 3)
Rhyolitic welded tuff (HyK-2-WR)	104.09	200	0.708992	±5	0.709090	±25	0.274	±47	(n = 2)
Carbonate precipitate samples									
Uwanari (pipe)	140.35	4,140	0.708543	±4	0.708581	±49	0.085	±50	(n = 2)
Uwanari (tank)	301.93	4,810	0.708532	±5	0.708552	±52	0.040	±47	(n = 2)
Ariake no.2 (tank)	444.40	594	0.708529	±5	0.708556	±49	0.060	±53	(n = 2)
Gokuraku (pipe)	221.02	596	0.708522	±5	0.708552	±53	0.061	±47	(n = 2)
Standards									
EN-1 (Carbonate, giant clam shell)	50.75		0.709174	±2	0.709268	±30	0.258	±27	(n = 6)
JCp-1 (Carbonate, coral)	29.12		0.709176	±3	0.709243	±14	0.194	±29	(n = 5)
BCR-1 (Basalt, Columbia River)	139.08		0.705020	±5	0.705105	±31	0.244	±35	(n = 4)
BHVO-2 (Basalt, Hawaii)	92.15		0.703481	±5	0.703567	±34	0.253	±37	(n = 4)
BIR-1a (Basalt, Iceland)	141.63		0.703105	±2	0.703200	±28	0.274	±35	(n = 4)
JB-1 (Basalt, Kitamatsuura)	116.31		0.704125	±2	0.704214	±30	0.257	±39	(n = 4)
JB-2 (Basalt, Oshima volcano)	102.20		0.703674	±3	0.703778	±31	0.293	±34	(n = 4)
JB-3 (Basalt, Fuji volcano)	116.48		0.703422	±7	0.703514	±37	0.266	±36	(n = 4)

Note. $^{87}\text{Sr}/^{86}\text{Sr}$: Normalized to $^{88}\text{Sr}/^{86}\text{Sr} = 8.375209$ and adjusted to $^{87}\text{Sr}/^{86}\text{Sr} = 0.710250$ of the NIST SRM987 Sr standard. $^{87}\text{Sr}/^{86}\text{Sr}^*$: Fractionated $^{87}\text{Sr}/^{86}\text{Sr}$ ratio from our double-spike correction algorithm. Strontium concentration determination used by an isotope dilution method. The forms of carbonate are identified using multipurpose X-ray diffraction (XRD) systems (a model Rigaku Ultima IV) at Kobe University.

^aUncertainties correspond to the last figures and are ± 2 standard errors (2se). ^bWeighted averages. Uncertainties (2s) correspond to the last figures. n : number of duplicated analyses of double-spiked samples.

In the Arima area, the $\delta^{88/86}\text{Sr}$ values of saline water samples from five sites range from 0.122 to 0.153‰, with an average of 0.141‰, and the strontium contents range from 15.4 to 37.7 $\mu\text{g g}^{-1}$. The average $^{87}\text{Sr}/^{86}\text{Sr}$ of saline water samples from Arima area is 0.708531 ± 0.000003 (2s, $n = 6$). The $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{88/86}\text{Sr}$ of the carbonate precipitates lie within the range of 0.70852–0.70854 and 0.040–0.085‰, respectively. The carbonate precipitates show more light-strontium enriched signatures than the saline water. Strontium contents in the calcite samples from Uwanari (4,100–4,800 $\mu\text{g g}^{-1}$) are seven times higher than those in aragonite samples from Gokuraku and Ariake ($\sim 600 \mu\text{g g}^{-1}$). The $\delta^{88/86}\text{Sr}$ of the Tansansen spring water is 0.248‰. The $\delta^{88/86}\text{Sr}$ values for the

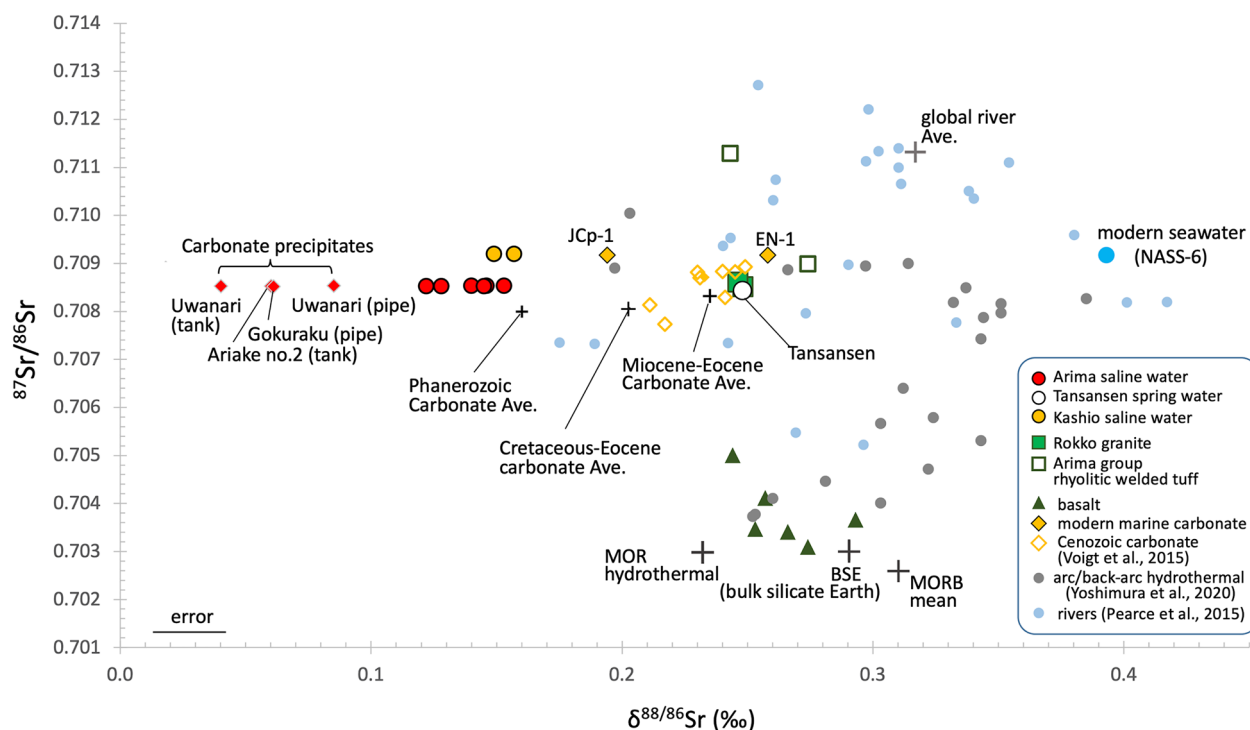


Figure 2. $^{87}\text{Sr}/^{86}\text{Sr}$ versus $\delta^{88/86}\text{Sr}$ plot of the Arima-type saline water, the Tansansen spring water, the carbonate precipitate samples, the Rokko granites and rhyolitic welded tuffs together with standard materials. Mean data of Phanerozoic, and Cretaceous-Eocene carbonates, global river water, mid-ocean ridge hydrothermal, mid-ocean ridge basalt (MORB), and the bulk silicate Earth (BSE) are from literatures (Vollstaedt et al., 2014; Voigt et al., 2015; Krabbenhöft et al., 2010; Caro & Bourdon, 2010; Amsellem et al., 2018; Pearce et al., 2015; Plank et al., 2007; Ito & Mahoney, 2005). Values on the axis of the ordinate are the traditional $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, thereby allowing comparison with previous data. Error bar is 2 standard error (2s).

Rokko granite (ROK-9: 0.248‰ and ROK-11: 0.246‰) and the Arima rhyolitic welded tuff (ROK-12: 0.243‰) samples are equivalent; however, at least one sample (HyK-2) displays a contrasting $^{87}\text{Sr}/^{86}\text{Sr}$ value.

In the Kashio area, the saline water samples from the two sites have an average of $\delta^{88/86}\text{Sr} = 0.153\text{‰}$ and show much higher strontium contents (82.3 and 162 $\mu\text{g g}^{-1}$). The $^{87}\text{Sr}/^{86}\text{Sr}$ of the saline water samples is 0.709203 ± 0.000002 (2s, $n = 2$), which is distinctively higher than that of the local basement, the Sanbagawa metamorphic rocks ($^{87}\text{Sr}/^{86}\text{Sr} < 0.7082$; Kawamoto et al., 2013; Ishikawa et al., 2014).

5. Discussion

5.1. Strontium Isotopic Characteristics of the Arima-Type Saline Water as Slab-Fluid Without Overprinting at Shallow Level

The $\delta^{88/86}\text{Sr}$ values obtained from the saline water are significantly lighter than those of modern seawater ($\delta^{88/86}\text{Sr}_{\text{NASS-6}} = 0.393\text{‰}$) and global rivers, and are also clearly different from those of local basement rocks (Figure 2). The $\delta^{88/86}\text{Sr}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ values of the Tansansen spring water, which are representative of the near-surface water, are almost identical to those of the Rokko granite (ROK-11) and thus likely reflect the shallow crust of this region. Whereas the $^{87}\text{Sr}/^{86}\text{Sr}$ value of the Arima saline water (0.70853) is only slightly higher than that of the Tansansen spring water (0.70844); the differing $\delta^{88/86}\text{Sr}$ values obviously distinguish the two (Figure 2). The strontium contents of the saline water (15.4–37.7 $\mu\text{g g}^{-1}$) are more than two orders of magnitude higher than that of the Tansansen spring water (0.147 $\mu\text{g g}^{-1}$). The $\delta^{88/86}\text{Sr}$ values suggest that the strontium isotopic composition of the Arima saline water is not overprinted by interaction with the local granite or welded tuff. The large differences of $^{87}\text{Sr}/^{86}\text{Sr}$ between the Kashio saline water and the basement rocks also indicate that the contribution from surrounding rocks to the saline water was minimal, if at all. Therefore, the $\delta^{88/86}\text{Sr}$ values of both saline water samples are not indicative of ambient $\delta^{88/86}\text{Sr}$ values at shallow level but should be regarded as specific characteristics of deep brine. The slab-fluid may escape to the surface without significant influence

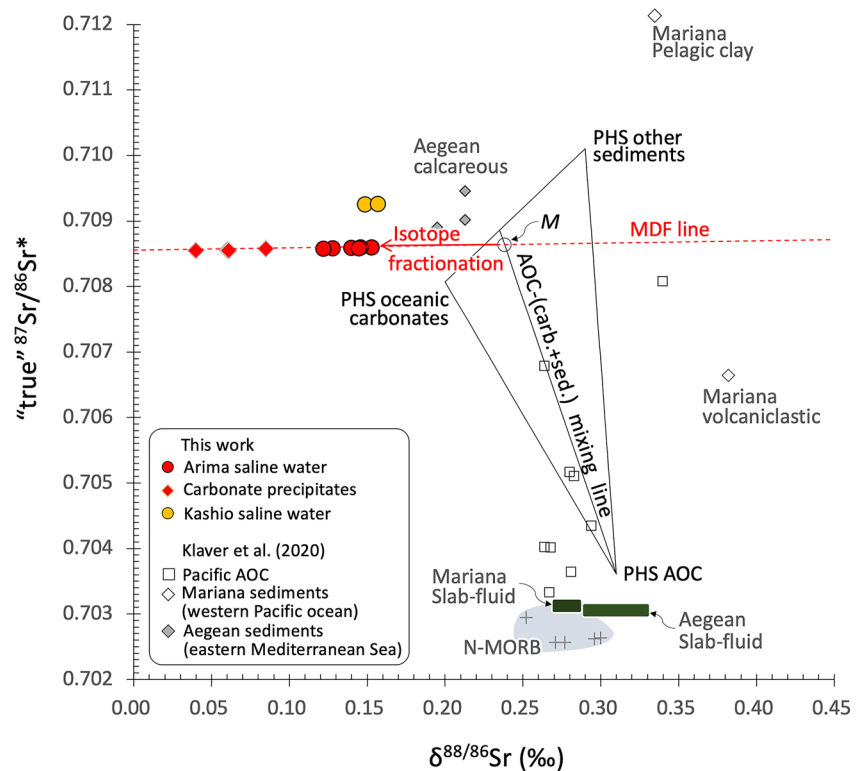


Figure 3. Strontium isotopic compositions of the Arima and Kashio saline water and their strontium sources on a “true” $^{87}\text{Sr}/^{86}\text{Sr}^*$ versus $\delta^{88/86}\text{Sr}$ plot. Mariana and Aegean sediments, Pacific AOC, and N-MORB are from Klaver et al. (2020). Mariana and Aegean slab-fluids were estimated from the Monte Carlo modeling (Klaver et al., 2020). Horizontal red dashed line: a MDF line through the $(^{87}\text{Sr}/^{86}\text{Sr}^*, \delta^{88/86}\text{Sr})_{\text{saline water}}$ with a slope of 0.00036 (Shalev et al., 2017), which intercepts a binary AOC-(carb. + sed.) mixing line at M ($^{87}\text{Sr}/^{86}\text{Sr}^* = 0.70861$, $\delta^{88/86}\text{Sr} = 0.239$)_{mixture}. The $(^{87}\text{Sr}/^{86}\text{Sr}^*, \delta^{88/86}\text{Sr})_{\text{saline water}}$ and the $(^{87}\text{Sr}/^{86}\text{Sr}^*, \delta^{88/86}\text{Sr})_{\text{carbonates}}$ values define an array [$^{87}\text{Sr}/^{86}\text{Sr}^* = (0.00034 \pm 0.00019) \times \delta^{88/86}\text{Sr} + (0.708538 \pm 0.000023)$] which corresponds to the MDF line. The $^{87}\text{Sr}/^{86}\text{Sr}^*$ ratio defines a mass-dependent relationship with $\delta^{88/86}\text{Sr}$ (for definition, see Appendix A of Krabbenhöft et al. (2010)). The $^{87}\text{Sr}/^{86}\text{Sr}^*$ of Kashio brine is more radiogenic than that of Arima brine, reflecting the differences in the age of the slab.

from rocks at crustal levels: the fluid-rock interaction at a high fluid/rock ratio in the long term would create a carbonate wall that prevent fluid-crustal rock interactions on the conduit/upwelling path guided by large faults.

Unlike other isotopes of volatile elements, the Arima-type saline water does not have magmatic/hydrothermal $\delta^{88/86}\text{Sr}$ values (Figure 2). The vent fluids from arc/back-arc seafloor systems of the Manus Basin, Izu-Bonin Arc, Mariana Trough, and western Pacific Fluids possess $\delta^{88/86}\text{Sr} > 0.25\text{‰}$, and even sediment-hosted hydrothermal fields in the Okinawa Trough, which interact with sedimentary carbonates, the $\delta^{88/86}\text{Sr}$ values are 0.20‰ (Yoshimura et al., 2020). The $\delta^{88/86}\text{Sr}$ values of the Arima-type saline water are exceptionally lighter than those values for river water and magmatic hydrothermal water. The $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{88/86}\text{Sr}$ have the potential to be used as an identifying isotopic index of slab-fluids, thereby quantitatively clarifying its spatial distribution, migration process, and water cycling in the subduction zone.

5.2. Light Strontium Isotope-Enriched Signature of Slab-Fluid

Modeling of slab-fluid from the subducted PHS plate is shown on a “true” $^{87}\text{Sr}/^{86}\text{Sr}^*$ versus $\delta^{88/86}\text{Sr}$ diagram (Figure 3). The following values were applied, for modeling: AOC attributes can be represented as $\delta^{88/86}\text{Sr} = 0.31\text{‰}$ for the average MORB (Amsellem et al., 2018), $^{87}\text{Sr}/^{86}\text{Sr} = 0.7035$ for the averages of oceanic basalts from neighboring region near along the Nankai Trough (West Philippine, Parece Vela, and Shikoku Basin basalts, Chen et al., 2021; Hickey-Vargas, 1991), and $\text{Sr} = 120 \mu\text{g g}^{-1}$ (Plank et al., 2007). Oceanic carbonates could be characterized as $\delta^{88/86}\text{Sr} = 0.20\text{‰}$ and $^{87}\text{Sr}/^{86}\text{Sr} = 0.7080$ for the Cretaceous-Eocene carbonates (Voigt et al., 2015; Vollstaedt et al., 2014), and $\text{Sr} = 330 \mu\text{g g}^{-1}$ for carbonate sediments (Krabbenhöft et al., 2010).

Other sediments can be represented as $\delta^{88/86}\text{Sr} = 0.29\text{‰}$ for bulk silicate Earth (Amsellem et al., 2018), $^{87}\text{Sr}/^{86}\text{Sr} = 0.7100$ for the Neogene bulk sediments (ODP site 1149) at Izu-Bonin (Plank et al., 2007), and $\text{Sr} = 165 \mu\text{g g}^{-1}$ for the bulk sediment composition at the Nankai Trough (Plank et al., 2007). Mass proportions of carbonates and other sediments from the Izu Trench sediments (44% carbonates and 56% other sediments; Plank et al., 2007) are required as a possible source. The $(^{87}\text{Sr}/^{86}\text{Sr}^*, \delta^{88/86}\text{Sr})_{44\text{carb.}+56\text{sed.}}$ and $(^{87}\text{Sr}/^{86}\text{Sr}^*, \delta^{88/86}\text{Sr})_{\text{AOC}}$ values define a mixing line between the two major sources of strontium in the PHS slab. The intercept of the binary mixing line and a mass dependent fractionation (MDF) line through the Arima saline water define the isotope composition of the AOC + sediments along with the mixing proportion of subducted material: 9% AOC, 40% carbonates and 51% other sediments. If this is the case, based on the difference between the Arima saline water and the unfractionated mixture, a fractionation factor between slab-fluid and the PHS slab ($\Delta^{88/86}\text{Sr}_{\text{fluid-slab}}$) is estimated to be -0.09 to -0.12‰ . The light-strontium enrichment in the Arima saline water is probably attributed to isotope fractionation during fluid generation. The newly estimated $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.70853 for the PHS slab is lower than previously thought (0.71087; Nakamura et al., 2008) and a contribution from the AOC to the slab-fluid is limited to $\leq 10\%$.

The slab-fluids released at forearc depth provide different insights into hydration/dehydration reactions than those obtained from arc lavas. Based on $^{87}\text{Sr}/^{86}\text{Sr}$ and $\delta^{88/86}\text{Sr}$ values of Mariana and Aegean arc lava, fluids were last in equilibrium with the lower oceanic crust at lower levels of the slab, suggesting that fluids at subarc depth originated from antigorite breakdown in the down-going lithospheric mantle and delivered to the mantle wedge (Klaver et al., 2020). In the southwest Japan arc, where the warm slab subducted, dehydration of blueschist, lawsonite, epidote, and amphibole in AOC occurs at forearc depth (Peacock & Wang, 1999), and the expelled aqueous fluids interact with the overlying oceanic sediments at the upper level of slab. Strontium which is concentrated in sediments/AOC (Plank & Langmuir, 1998) migrates to the mantle wedge together with the fluids.

According to an experimental study (Kessel et al., 2005), the partitioning coefficient between aqueous fluid and solid ($D^{\text{fluid/solid}}$) of strontium at 4 GPa is ~ 0.3 at 700°C and is greater than unity at $\geq 800^\circ\text{C}$. Because the pressure-temperature (P - T) conditions for slab-fluid generation beneath the Arima area were 1.5–2.4 GPa and 400 – 500°C (Kusuda et al., 2014), the $D^{\text{fluid/solid}}$ of strontium at the P - T conditions is expected to be much smaller than unity. If isotopic fractionation occurred under the condition of the low mobility of strontium during fluid generation, the influence of kinetic effects might result in enrichment in isotopically light strontium in the fluids. While isotope fractionation may not be prominent in high-temperature processes such as magmatic differentiation and fractional crystallization (Charlier et al., 2012), strontium isotope fractionation could have occurred in fluid-rock interactions at lower temperatures at forearc depth during dehydration reactions. The light isotope enrichment in fluids generated at forearc depth may be explained by the small $D^{\text{fluid/solid}}$ of strontium at low temperature and pressure within forearc compared to subarc.

5.3. Comparison Between Arima and Kashio Brines

The Arima and Kashio areas are ~ 270 km apart and their basement rocks; that is, the Rokko granites with respect to Arima and the Sanbagawa metamorphic rocks at Kashio are completely different. Nevertheless, the similarity of the $\delta^{88/86}\text{Sr}$ values of the saline water from both sites implies commonality of their origin. The similarity of the source materials to both brines is recognized based on the non-meteoric hydrogen and oxygen isotopic compositions and mantle-derived $^3\text{He}/^4\text{He}$, while the $\delta^{18}\text{O}$ and δD values and chlorine contents in the Kashio brine are lower than those in the Arima brine, suggesting a certain variability in the composition of slab-fluid because of the differences in the depth of dehydration (Kusuhara et al., 2020). The $^{87}\text{Sr}/^{86}\text{Sr}^*$ of the Kashio brine (0.70925) is more radiogenic than that of the Arima brine (0.70858), which may reflect variation in the fluid compositions caused by differences in the age of the slabs (i.e., the young PHS slab and the older PAC slab).

5.4. Strontium Isotope Fractionation During Precipitation of Carbonates From the Arima Spring Water

On Figure 3 data points for the Arima saline water and carbonate precipitates yield a linear array which corresponds to a MDF line. The carbonates are light strontium enriched relative to their precipitation saline water, and a fractionation factor $\Delta^{88/86}\text{Sr}_{\text{carb. precipitates-saline water}}$ is approximately -0.10‰ . The fractionation is smaller than that between skeletal carbonates and seawater ($\Delta^{88/86}\text{Sr}_{\text{carb.-seawater}} = -0.21\text{‰}$; Vollstaedt et al., 2014). Strontium isotope fractionation of inorganic calcite is dominantly controlled by the precipitation rate (increasing

fractionation with higher rates), which is due to chemical kinetic fractionation of the hydrated cations (preferred desolvation of the isotopically light cations) (Böhm et al., 2012). The carbonate growth rate in the Arima saline water ($\sim 1.6 \times 10^6 \mu\text{mol m}^{-2} \text{hr}^{-1}$) is larger than the calcification of foraminiferal calcite ($10^4 \mu\text{mol m}^{-2} \text{hr}^{-1}$; Böhm et al., 2012). A fractionation factor $\Delta^{88/86}\text{Sr}_{\text{carb. precipitates-saline water}}$ may be controlled by temperature and/or processes at a diffusive boundary layer.

6. Conclusions

The first application of stable strontium isotopes for the Arima-type saline water successfully demonstrates that the saline water sampling from both the Arima and Kashio areas possesses light strontium-rich signatures ($\delta^{88/86}\text{Sr} = 0.122\text{--}0.157\text{‰}$). A possible interpretation of the light strontium enrichment in the Arima-type saline water is the consequences of isotope fractionation during fluid generation from the subducted AOC and sediments rather than deriving from local rocks or near-surface water. If it is given as strontium isotope fractionation signatures of slab-fluid released at forearc depth ($\Delta^{88/86}\text{Sr}_{\text{fluid-slab}} = -0.10\text{‰}$), $\delta^{88/86}\text{Sr}$ of the Arima-type brine could be utilized as a new index of slab-fluids for further evaluation of fluid formation and water/element cycling in the subduction-zones.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

The data used in this study are available through the Arctic Data archive System (ADS) (Kani et al., 2022). The ADS is operated by Polar Environment Data Science Center, National Institute of Polar Research.

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