A Systematic Assessment of Stable Sr Isotopic Compositions of Vent Fluids in Arc/Back-Arc Hydrothermal Systems: Effects of Host Rock Type, Phase Separation, and Overlying Sediment

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Variations in the stable isotopic composition of seawater Sr (δ88Sr) is a new tool for estimating the rates of global carbonate sedimentation over geologic time, yet the isotope compositions of the major sources and sinks of Sr to the world oceans are still in need of further constraint. We report δ88Sr values of vent fluids from arc/back-arc seafloor hydrothermal systems in the western Pacific. In the sediment-starved hydrothermal fields of the Manus Basin, Izu-Bonin Arc, and Mariana Trough, the δ88Sr values of end-member fluids for each site showed little variation (0.29–0.30‰) and were close to the average value of oceanic volcanic rocks, reflecting dissolved Sr sourced from host rocks. Chlorine-depleted fluids from phase-separated hydrothermal systems in the North Fiji Basin had the end-member δ88Sr values of 0.26, 0.28, and 0.29‰. Thus, both sediment-starved and phase-separated vent fluids had the end-member δ88Sr values indistinguishable from or very close to the range of oceanic volcanic rocks. Therefore, the δ88Sr compositions in these hydrothermal sites are controlled predominantly by Sr sourced from host rock with a small influence from secondary mineral precipitation/re-dissolution. Fluids from the sediment-hosted hydrothermal fields of the Okinawa Trough, however, were characterized by low δ88Sr values of approximately 0.22‰ and high 87Sr/86Sr ratios, indicating interactions with sedimentary carbonates. As for the modern oceanic δ88Sr budget, the sediment-hosted sites lower the global hydrothermal δ88Sr. Since both sediment-starved and -hosted hydrothermal systems provide a long-term control on the global Sr cycle, the end-member δ88Sr value is an important constraint on the evolution of Sr cycling in past oceans.

Keywords: strontium, isotopes, arc, back-arc, convergent margin, hydrothermal vent fluid
INTRODUCTION

Seafloor hydrothermal systems play an important role in chemical and thermal fluxes to the ocean, which impact the evolution of ocean chemistry, climate, and life throughout geologic time (Elderfield and Schultz, 1996; Hardie, 1996). The concentration of $\text{Sr}^{2+}$ in seawater is 89 nM, the fifth most abundant cation in seawater, with a mean residence time of about 2.5 million years (Hodell et al., 1990). Strontium inputs from hydrothermal systems have played an important role in past changes of seawater $\delta^{88}\text{Sr}$/$\delta^{86}\text{Sr}$ values, accounting for approximately 5% of total input fluxes with end-member $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ values of 0.703–0.704 (Pearce et al., 2015a). These changes in the $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ value of seawater have been used to elucidate the relative intensities of continental weathering and hydrothermal alteration of oceanic crust over time (DePaolo and Ingram, 1985; Hodell et al., 1990; Raymo and Ruddiman, 1992; Veizer et al., 1999; McArthur et al., 2001).

The combined use of radiogenic ($\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$) and stable isotopic ratios ($\delta^{88}\text{Sr}$/$\delta^{86}\text{Sr}$, denoted by $\delta^{88}\text{Sr}$) is a novel approach that is useful for identifying differences in Sr sources and processes involving chemical reactions in the Earth’s surface (Figure 1; Ohno and Hirata, 2007; Halicz et al., 2008; Ohno et al., 2008; Wei et al., 2013; Neymark et al., 2014; Chao et al., 2015; Andrews et al., 2016; Stevenson et al., 2016; Liu et al., 2017; Shalev et al., 2017). Fractionation of $\delta^{88}\text{Sr}$/$\delta^{86}\text{Sr}$, but not of $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$, accompanies mineral precipitation and biological uptake (Böhm et al., 2012; Stevenson et al., 2014), and is of importance in understanding isotopic mass balance in Earth’s Sr cycle. The utility of the isotopic mass balance of $\delta^{88}\text{Sr}$ and $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ for the marine Sr budget has been favorably assessed in studies of both modern and past oceans (Krabbenhöft et al., 2010; Pearce et al., 2015a).

Modern seawater is isotopically quite homogeneous in terms of both $\delta^{88}\text{Sr}$ and $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$, irrespective of area or depth (0.39‰ and 0.70917, Wakaki et al., 2017), and seawater Sr concentrations reflect the balance between contributions from hydrothermal systems, marine diagenesis, chemical weathering of continental rocks to seawater, and carbonate burial. The evolution of the seawater $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ value is determined by the balance of inputs from rivers (approximately 0.713) and hydrothermal systems (0.704), whereas the $\delta^{88}\text{Sr}$ record reflects primarily changes in the balance between riverine (0.32‰) and hydrothermal (0.24–0.27‰) inputs (Figure 1). The removal of marine biogenic carbonates 0.18–0.24‰ depleted in $86\text{Sr}$ leaves the ocean isotopically heavier than $\delta^{88}\text{Sr}$ values of the input fluxes (Krabbenhöft et al., 2010; Pearce et al., 2015a). As a result, $\delta^{88}\text{Sr}$ of past seawater varied from 0.25 to 0.60‰ in the Phanerozoic era (Vollstedt et al., 2014). Strontium inputs from hydrothermal systems play a major role in the $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ flux to the ocean (Elderfield and Schultz, 1996), but the magnitude of Sr isotope fractionation and the compositional variation of the end-member solutions in hydrothermal water-rock reactions must be known for accurate estimation of the global marine Sr input and output fluxes (Krabbenhöft et al., 2010). Strontium uptake and release during oceanic hydrothermal circulation may result in its enrichment in certain isotopes from the recharging seawater; the resolution of this key issue requires further investigation in different types of hydrothermal settings.

Most data reported from Mid-Atlantic Ridge (MAR) hydrothermal vent fluids (Krabbenhöft et al., 2010; Pearce et al., 2015a) lie on the mixing line between seawater and an end-member component with estimated $\delta^{88}\text{Sr}$ and $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ values of 0.27‰ and 0.7045, respectively (Krabbenhöft et al., 2010), or 0.24‰ and 0.7037 (Pearce et al., 2015a), respectively. These end-member values are consistent with the reference values of oceanic basalts (about 0.25‰, Halicz et al., 2008; Ohno et al., 2008; Pearce et al., 2015a), although the $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ values are slightly higher than those of fresh mid-ocean ridge basalt (MORB). More recently, the $\delta^{88}\text{Sr}$ value of MORB has been suggested to be 0.31 ± 0.05‰ (Amsellem et al., 2018). In some MAR hydrothermal fluids, however, $\delta^{88}\text{Sr}$ values are higher than expected from the mixing trend (Figure 2), and these higher values are interpreted as a result of isotopic fractionation during hydrothermal mineral precipitation or dissolution (Pearce et al., 2015a). A recent experimental study conducted under high-temperature and high-pressure conditions has shown that in fluid-basalt interactions, $88\text{Sr}$ is preferentially taken up into anhydrite (i.e., greater anhydrite precipitation and residual Sr dissolution in lower and higher $\delta^{88}\text{Sr}$ in the fluid, respectively) and $86\text{Sr}$ is preferentially released as a result of the incongruent dissolution of crystalline basalt (Voigt et al., 2018). It is still uncertain to what degree the variation of $\delta^{88}\text{Sr}$ values in hydrothermal systems can be attributed to solute sources or secondary mineral (e.g., anhydrite and calcite veins) formation.

In arc and back-arc basins, the chemical compositions of vent fluids vary more widely than those of vent fluids in sediment-starved hydrothermal sites because of the compositional variation of the host rocks as some are buried by sediments (Volpe et al., 1987). Given the tectonic differences between back-arc rifting and mid-ocean ridge spreading zones, it is essential to fully assess $\delta^{88}\text{Sr}$ systematics in arc/back-arc settings. In addition, although phase separation and segregation of vapor and fluid phases greatly affect hydrothermal fluid chemistry in both mid-ocean ridge and arc/back arc settings (Volpe and Bischoff, 1987), the impact of separation and segregation processes on the $\delta^{88}\text{Sr}$ values of hydrothermal fluids has not yet been examined. Finally, as an extreme case, hydrothermal fields of back-arc setting include heavily sediment-covered rift that lacks typical oceanic crust, as represented by the Okinawa Trough (Figure 3). Terrigenous materials contained in the overlying sediments have considerably influenced the $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ signatures of the hydrothermal fluids in this region (Chiba et al., 1993; Noguchi et al., 2009; Araoka et al., 2016), and $\delta^{88}\text{Sr}$ can be utilized to precisely apportion Sr sources in these sediment-hosted fields, that also impact the oceanic $\delta^{88}\text{Sr}$ inventory.

The characteristics of hydrothermal systems in arc/back arc settings are diverse with respect to the presence or absence of marine sediments, host-rock type, phase separation, and temperature of the vent fluids. However, how these factors influence $\delta^{88}\text{Sr}$/$\delta^{86}\text{Sr}$ fractionation and variations in the $\delta^{88}\text{Sr}$ values of the fluids remains unknown. In this study, we investigated the $\delta^{87}\text{Sr}$/$\delta^{86}\text{Sr}$ and $\delta^{88}\text{Sr}$ systematics of
hydrothermal fluids collected from 11 vents in five arc and back-arc hydrothermal systems in the western Pacific. The studied sites, which include both sediment-starved and sediment-hosted sites and cover the large variation in hydrothermal settings described above, provide essential information for comprehensive understanding of Sr geochemistry in hydrothermal systems.

MATERIALS AND METHODS

Geological Settings of Hydrothermal Vents

Hydrothermal vent fluids previously collected from eight sediment-starved sites in the Manus Basin (n = 5), Izu-Bonin Arc (n = 3), Mariana Trough (n = 4), and North Fiji Basin (n = 6), and from three sediment-hosted sites in the Okinawa Trough (n = 7) are used for this study (Table 1; Figure 3) (Sakai et al., 1990a; Gamo et al., 1991; Grimaud et al., 1991; Chiba et al., 1993; Ishibashi, 1994; Ishibashi et al., 1994a; Gamo et al., 1997). The chemical variability of the vent fluids as well as hydrothermal mineral deposits of these arc/back-arc hydrothermal systems reflect diversity in geological structures (axial graben, crest of a ridge, seamount, flank of an axial volcano, volcanic depression, and sediment-rich depression) and lithologic types ranging from mafic to felsic rocks (Ishibashi and Urabe, 1995; Table 2). The sediment-hosted Okinawa Trough sites are all developed on continental lithosphere, not oceanic lithosphere. According to the estimate of hydrothermal fluid fluxes from the major types of hydrothermal systems (Nakamura and Takai, 2015), the sediment-associated hydrothermal systems yield approximately 10% of the total hydrothermal flux to seawater; the sediment-starved mid-ocean ridge and arc/back-arc hydrothermal systems are estimated to deliver 80% and 10%, respectively. Chemical properties of sediment-starved hydrothermal system are controlled essentially by chemical equilibria between fluids and host rocks, while the hydrothermal fluids in the Okinawa Trough have been shown to interact with terrigenous and biogenic materials (Sakai et al., 1990a; Gamo et al., 1991).

Sediment-Starved Sites (Manus Basin, Izu-Bonin Arc, Mariana Trough, North Fiji Basin)

The Vienna Woods field is located on the central spreading axis in the Manus Basin. This site is hosted in basalt and the chemical characteristics of the vent fluids have been previously reported to be comparable to those of mid-ocean ridge hydrothermal fluids (Lisitsyn et al., 1993; Gamo et al., 1997). The vent fluids of the Pacmanus field, located in the Eastern Manus Volcanic Zone, are characterized by higher K and B concentrations and a lower Ca
concentration compared with the Vienna Woods fluids, reflecting their interaction with dacitic host rocks (Yamaoka et al., 2015).

The Suiyo Seamount is a dacitic submarine volcano along the volcanic front of the Izu-Bonin Arc. Its fluids are enriched in Ca and Sr, as well as in gaseous CO₂, compared with the fluids from the mid-ocean ridge site (Ishibashi, 1994; Tsunogai et al., 1994).

The Mariana Trough is an actively spreading back-arc basin behind the Mariana Arc. The Alice Springs field is located on the flank of an axial basaltic andesite volcano in the mid-Mariana Trough. Clear, low-temperature (202°C) vent fluids were collected from an anhydrite chimney in the Forecast Vent field at the summit of a seamount in the southern Mariana Trough (Gamo et al., 1994).

The White Lady, Kaiyo, and LHOS sites in the North Fiji Basin have low chlorinity, suggesting segregation of vapor-rich phases (Grimaud et al., 1991). The petrological characteristics of the basalt in the North Fiji Basin are similar to those of normal MORB (N-MORB) but include a regional influence of coexisting oceanic island basalt components (Eissen et al., 1991; Nohara et al., 1994). Fluid samples from the White Lady field were collected from an anhydrite chimney at a topographic high point in the axial graben of the northern-central segment near the triple junction. Two chimneys, Kaiyo and LHOS, are also located in the same segment, 150 m southwest of the White Lady field. The chlorinity of vent fluids in the North Fiji Basin is estimated to be 47–49% that of seawater, suggesting that phase separation occurs beneath the seafloor and that a vapor-rich phase has condensed to form the hydrothermal fluid (Grimaud et al., 1991; Ishibashi et al., 1994b).

Sediment-Hosted Sites (Okinawa Trough)

The Okinawa Trough is a back-arc basin located at the eastern margin of the Eurasian Continent. It is considered to be initiated as continental rifting which commenced since approximately 2 Ma (Sibuet et al., 1995). Hydrothermal sites in the Okinawa Trough are hosted in basalt for JADE and rhyolite-andesite for CLAM sites, and accompanied by a thick sediment layer consisting mainly of terrigenous clay derived from the Eurasian Continent (e.g., the Yangtze and Yellow Rivers) and biogenic carbonates (Halbach et al., 2017). For hydrothermal sites in the mid-ocean ridges, the primary fluid source is overlying...
seawater recharged through basaltic outcrops. In contrast, the large reservoir of sedimentary pore water in the thick trough-fill sediments recharges hydrothermal fluids in the sediment-hosted hydrothermal system of the mid-Okinawa Trough (Figure 4; Tsuji et al., 2012). Fluid migration driven by thermal advection of the silicic arc volcanism is considered to flow laterally and updip along permeable volcanoclastic sedimentary layers and along basement faults within the crust (Tsuji et al., 2012). These sediment-hosted hydrothermal systems are highly alkaline and enriched in methane derived from thermally altered sedimentary organic matter (Von Damm et al., 1985; Gamo et al., 1991; Kawagucci et al., 2013). The hydrothermal fluids in this area have high alkalinity and are enriched in NH$_4^+$, CH$_4$, and CO$_2$ as a result of interactions with sediments (Sakai et al., 1990a; Sakai et al., 1990b; Gamo et al., 1991; Chiba et al., 1993; Kawagucci et al., 2013).

### TABLE 1 | Sr isotopic ratios and other chemical characteristics of hydrothermal fluid and seawater samples.

| Site          | Sr (μmol/kg) | Sr/Ca (mmol/mol) | $^{87}$Sr/$^{86}$Sr | $\delta^{88}$Sr (‰) | 2sd | Mg (mmol/kg) | Ca (mmol/kg) | SO$_4$ (mmol/kg) | Cl (mmol/kg) |
|---------------|--------------|------------------|---------------------|---------------------|-----|--------------|--------------|----------------|--------------|
| Sediment-starved sites |
| 303-TS1 Vienna woods | 115 | 6.5 | 0.707972 | 0.351 | 0.025 | 46.3 | 17.6 | 25.3 | 565 |
| 307-1 Vienna woods | 187 | 3.7 | 0.705312 | 0.343 | 0.021 | 20.3 | 50.6 | 11.6 | 629 |
| 307-2 Vienna woods | 256 | 3.4 | 0.704465 | 0.281 | 0.025 | 2.5 | 74.7 | 2.0 | 687 |
| 301-8 Pacmanus | 114 | 8.5 | 0.706625 | — | — | 21.0 | 13.4 | 11.3 | 564 |
| 304-1 Pacmanus | 88 | 9.2 | 0.708495 | 0.337 | 0.025 | 42.0 | 9.5 | 20.8 | 545 |
| D630-3 Suiyo seamount | 270 | — | 0.704178 | 0.322 | 0.025 | 17.7 | — | — | — |
| D651-2 Suiyo seamount | 307 | — | 0.704011 | 0.303 | 0.025 | 0.8 | — | — | — |
| D651-6 Suiyo seamount | 295 | — | 0.704109 | 0.260 | 0.022 | 3.7 | — | — | — |
| D154-2 Alice springs | 84 | 3.9 | 0.703737 | 0.252 | 0.025 | 0.9 | 21.7 | D.L. | 582 |
| D154-5 Alice springs | 94 | 4.2 | 0.703774 | 0.253 | 0.028 | 1.0 | 22.3 | D.L. | 548 |
| D182-4 Forecast vent | 101 | 6.0 | 0.707876 | 0.344 | 0.021 | 43.2 | 16.8 | — | — |
| D187-2 Forecast vent | 147 | 4.0 | 0.705794 | 0.324 | 0.026 | 25.3 | 36.5 | — | — |
| Phase-separated sites |
| IST5-2 White lady | 42 | 5.5 | 0.706405 | 0.312 | 0.025 | 8.6 | 7.7 | 4.9 | 302 |
| IST6-1 White lady | 66 | 8.0 | 0.708164 | 0.351 | 0.025 | 25.7 | 8.3 | 13.3 | 402 |
| D80-3 Kayo | 79 | 8.9 | 0.708273 | 0.385 | 0.026 | 31.5 | 8.9 | 16.7 | 420 |
| D80-4 Kayo | 60 | 6.7 | 0.705668 | 0.303 | 0.015 | 8.5 | 9.0 | 4.5 | 318 |
| D93-3 LHOS | 77 | 7.7 | 0.707433 | 0.343 | 0.012 | 24.4 | 10.0 | 13.3 | 401 |
| D93-4 LHOS | 80 | 8.2 | 0.708194 | 0.332 | 0.021 | 33.5 | 9.7 | 18.1 | 452 |
| Sediment-hosted sites |
| D423-3 JADE | 180 | 4.5 | 0.708911 | 0.197 | 0.018 | 1.0 | 40.0 | D.L. | 528 |
| D423-5 JADE | 100 | 7.1 | 0.709152 | — | — | 45.2 | 14.1 | 24.1 | 503 |
| D621-5 Minami-ensei | 90 | 5.4 | 0.708875 | 0.266 | 0.013 | 27.2 | 16.7 | 13.7 | 550 |
| D622-1 Minami-ensei | 247 | 10.8 | 0.710024 | — | — | 4.9 | 22.9 | D.L. | 550 |
| D622-2 Minami-ensei | 252 | 10.9 | 0.710051 | — | — | 1.4 | 23.2 | D.L. | 550 |
| D426-6 CLAM | 74 | 5.2 | 0.709000 | 0.314 | 0.016 | 39.5 | 14.3 | 20.7 | — |
| D427-6 CLAM | 91 | 4.7 | 0.708948 | 0.297 | 0.013 | 33.4 | 19.2 | — | 517 |
| Seawater | — | 89 | 8.7 | 0.709172 | 0.392 | 0.025 | 52.7 | 10.3 | 28.0 | 546 |

### TABLE 2 | Characteristics of seafloor hydrothermal vents and Sr isotopic end-member compositions.

| Site          | Type | Host rock | Temp. (°C) | pH at 25°C | Sr end-members |
|---------------|------|-----------|------------|------------|----------------|
| Sediment-starved sites |
| Vienna woods | Back arc | Basalt | 302 | 4.0–4.5 | 260 | 0.70447 | 0.298 |
| Pacmanus | Back arc | Dacite | 288 | 2.5 | 126 | 0.70583 | — |
| Suiyo seamount | Arc | Dacite | 311 | 3.7 | 320 | 0.70407 | 0.291 |
| Alice springs | Back arc | Basalt | 280 | 3.9 | 89 | — | — |
| Forecast vent | Back arc | Basalt | 202 | — | 196 | 0.70440 | 0.288 |
| Phase-separated sites |
| White lady | Back arc | Basalt | 285 | 4.7 | 36 | 0.70546 | 0.280 |
| Kayo | Back arc | Basalt | 291 | 4.7 | 56 | 0.70495 | 0.290 |
| LHOS | Back arc | Basalt | 291 | 4.7 | 66 | 0.70556 | 0.256 |
| Sediment-hosted sites |
| JADE | Back arc | Rhyolite-andesite | 320 | 4.7 | 181 | 0.70892 | 0.195 |
| Minami-ensei | Back arc | Basalt | 278 | 4.9–5.1 | 190 | 0.70988 | 0.235 |
| CLAM | Back arc | Basalt | 220 | 5.3 | 74 | 0.70909 | — |
The JADE site is a high-temperature (320°C) hydrothermal vent field in the middle Okinawa Trough, and the fluids are emitted from sulfide-sulfate chimneys. The Minami-Ensei knoll is located in a rift center in the northern Okinawa Trough (Chiba et al., 1993). This knoll is a volcanic complex characterized by a complicated topography of small hills and depressions within a circular rim of 2 km radius. The samples were collected from fragile anhydrite chimneys. The CLAM hydrothermal field is located in a small depression along the eastern Ihey Ridge and is associated with basalt breccia (Ishibashi et al., 1995). Mineralized chimney precipitates of the CLAM field consist mainly of manganese calcite, rhodochrosite, anhydrite, and amorphous silica (Izawa et al., 1991). At the CLAM site, the end-member fluids contain Mg$^{2+}$ (approximately 20 mM), reflecting a low maximum temperature of 220 °C and suggesting an incomplete water-rock reaction (Sakai et al., 1990b).

**Analytical Methods**

Hydrothermal fluid samples were filtered, acidified with HNO$_3$, and stored in polypropylene bottles. Major and trace element concentrations and δ$^7$Li, δ$^{11}$B, δ$^{26}$Mg, 87Sr/86Sr values of the fluid samples have been reported previously (Yamaoka et al., 2015; Araoka et al., 2016; Eom et al., 2020).

δ$^{88}$Sr and 87Sr/86Sr values of the samples were determined simultaneously using the double-spike technique (Wakaki et al., 2017) in a TRITON thermal ionization mass spectrometer (Thermo Scientific, Germany) at Kochi Core Center, Japan. Two aliquots of each fluid sample, with and without the addition of the 84Sr–86Sr spike, were prepared for Sr isotopic analysis. After chemical separation of Sr by a Sr-Spec resin (Eichrom resin), the Sr sample with a Ta activator was loaded onto a single W filament. The 88Sr beam intensities were approximately 11 and 4 V for the unspiked and spiked runs, respectively. Peaks for 86Sr, 87Sr, 88Sr, 84Sr, and 85Rb were monitored. The 87Sr/86Sr ratio was measured in the unspiked sample by the conventional internal normalization technique using 86Sr/88Sr = 0.1194 and the exponential law. Stable 88Sr/86Sr ratios were calculated by using the data from the unspiked and spiked runs, and expressed as permil (%o) deviations relative to the NBS-987 standard as:

$$\delta^{88}Sr = \left( \frac{^{88}Sr/^{86}Sr}_{\text{sample}} / \frac{^{88}Sr/^{86}Sr}_{\text{NBS-987}} - 1 \right) \times 10,000.$$  (1)

Repeated analyses of the in-house Sr isotopic reference reagent Wako-9999 (Wakaki et al., 2017) gave δ$^{88}$Sr = 0.322 ± 0.025‰ (2 SD, n = 45). The external error for vent fluid samples was estimated as the 2SD repeatability of multiple SRM-987 analyses during each analytical session (Wakaki et al., 2017) and ranged from ±0.013‰ to ±0.028‰ (Table 1). To check the accuracy of the analysis, we analyzed the IAPSO seawater (0.392 ± 0.015‰, 2 SE, n = 3) as reference material, and consistent with published values for seawater samples from the Atlantic, Pacific, Indian, and Southern Oceans (Krabbenhöft et al., 2009; Neymark et al., 2014; Stevenson et al., 2014; Vollstaedt et al., 2014; Pearce et al., 2015a). The conventional 86Sr/88Sr ratios obtained by unspiked runs were corrected for a small systematic mass bias to give 87Sr/86Sr$_{\text{SRM-987}}$ = 0.710248. Typical repeatability of the 86Sr/88Sr ratios was ±0.000005 (2SD). The 87Sr/86Sr values determined in this study were essentially consistent with those previously reported by Araoka et al. (2016). For consistency with the δ$^{88}$Sr values, we used the newly determined 87Sr/86Sr values in the present analysis.

**RESULTS**

We examined the relationship between δ$^{88}$Sr and 87Sr/86Sr in the hydrothermal fluids and compared the result with data for MAR hydrothermal fluids (Krabbenhöft et al., 2010; Pearce et al., 2015a) and MORB (Amsellem et al., 2018) (Figure 2; Table 1). The samples from sediment-starved (Manus Basin, Izu-Bonin Arc, and Mariana Basin) and phase-separated (North Fiji Basin) sites showed a clear positive correlation between δ$^{88}$Sr and 87Sr/86Sr (Figure 2) and plot right along with the data reported previously by Krabbenhöft et al. (2010) for MAR. All 87Sr/86Sr values of the fluids from the sediment-hosted sites (Okinawa Trough) were distinctly higher than those from the other sites, and the δ$^{88}$Sr values were distinctly lower. The fluids from sites in the Okinawa Trough had the following characteristics: 1) their 87Sr/86Sr values are similar to or even higher than the seawater value; 2) on the 87Sr/86Sr–δ$^{88}$Sr scatter plot, they are outliers compared with the trend for sediment-starved sites; and 3) δ$^{88}$Sr values of fluids from the JADE and...
Minami-Ensei sites were approximately 0.20‰ or less (Figure 2; Table 1).

Because hydrothermal fluid samples represent a mixture of seawater and the end-member hydrothermal fluid, previous studies calculated δ⁸⁸Sr end-member values by extrapolating Mg/Sr to zero (Supplementary Figure S1). This approach is based on the hypothesis that Sr and Mg concentrations are closely coupled through water-rock reactions. However, hydrothermal fluids can experience either a net gain or loss of Sr, compared with the initial seawater concentration, during hydrothermal fluid circulation (Kawagucci et al., 2013), irrespective of whether the Mg concentration is close to zero in the end-member fluid. In this study, the Sr concentrations of the end-member fluid at each site were estimated by extrapolating the observed Sr-Mg linear regression line fitted to the data of two or three samples from each site to Mg = 0 (Figures 5A–E). For the CLAM site only, the regression line was extrapolated to an estimated end-member Mg content of 20 mM (Araoka et al., 2016) based on the observed relatively low maximum temperature of approximately 220 °C (Sakai et al., 1990a; Gamo et al., 1991).
At most sites, both $^{87}$Sr/$^{86}$Sr (Figures 5F–J) and $^{88}$Sr (Figures 5K–O) values were linearly related to 1/Sr; therefore, the end-member Sr isotopic compositions were calculated using the end-member Sr concentrations. All linear regression lines were forced through the seawater end point. Significant correlations were not observed between $^{88}$Sr and 1/Sr in samples from the Pacmanus, Alice Springs, and CLAM sites because these samples had a Sr concentration similar to that of seawater. The estimated end-member compositions are summarized in Table 2 along with major oceanic fluid components reported by Pearce et al. (2015a). Compared to seawater values ($^{87}$Sr = 89 μmol/kg, $^{88}$Sr = 0.392‰, and $^{86}$Sr/$^{88}$Sr = 0.709, 172), the end-member fluids of the sediment-starved sites were characterized by higher Sr concentrations of up to 320 μmol/kg and lower $^{88}$Sr and $^{86}$Sr/$^{88}$Sr values of 0.29–0.30‰ and 0.7041–0.7058, respectively. Slight differences from the $^{87}$Sr/$^{86}$Sr values reported by Araoka et al. (2016) are because the regression line was forced through the seawater end point. Most Sr isotopic end-member values for samples from the phase-separated sites were similar to those of samples from the sediment-starved sites, but the Sr concentrations were notably lower. The $^{88}$Sr end-member of most sediment-starved sites is in good agreement with MORB composition. The global marine $^{88}$Sr and $^{86}$Sr/$^{88}$Sr fluxes are assessed by an intercept point between 1) a fractionation line between seawater-carbonate and 2) a binary mixing line of hydrothermal-river endmembers (Figure 6; Krabbenhöft et al., 2010; Pearce et al., 2015a). The LHOS $^{88}$Sr value (0.26‰) was lower than the values at the sediment-starved and other phase-separated sites. The sediment-hosted sites in the Okinawa Trough exhibited markedly high $^{87}$Sr/$^{86}$Sr (0.7089–0.7099) and low $^{88}$Sr (0.20–0.24‰) values, and Sr concentrations varied greatly among these sites (Table 2). The end-member values for the sediment-hosted sites are close to the value of marine biogenic carbonates ($^{88}$Sr = 0.150‰, $^{87}$Sr/$^{86}$Sr = 0.70917) rather than that of igneous rocks (Figure 6).

**DISCUSSION**

**Host Rock and Fractionation Controls on $^{88}$Sr in Sediment-Starved Systems**

The data obtained in this study provide insights into the control of $^{88}$Sr in end-member fluids associated with hydrothermal systems with varied host rock compositions and with or without phase separation. The Vienna Woods field in the Manus Basin is a basalt-hosted hydrothermal system in MORB-like basaltic rocks (Dril et al., 1997; Gamo et al., 1997). In contrast, the host rocks of the Pacmanus field in the Manus Basin and the Suiyo Seamount site in the Izu-Bonin Arc are arc-type dacitic rocks. The geochemical characteristics of the Mariana Trough basalts reflect a mixing of melts in the source region prior to eruption (Volpe et al., 1987) with abundant CO$_2$ derived from subducting slab material (Gamo et al., 1997). Among these sites, the dacite-hosted Pacmanus site alone has a high $^{87}$Sr/$^{86}$Sr value of 0.7058. The $^{88}$Sr end-member values at the five sediment-starved sites in these hydrothermal fields are similar at approximately 0.29‰ (Table 2). The estimated $^{88}$Sr end-member values are 0.02–0.04‰ higher than those of the MAR reported by Krabbenhöft et al. (2010) and Pearce et al. (2015a). Amsellem et al. (2018) recently demonstrated that igneous processes have a limited effect on $^{88}$Sr/$^{86}$Sr value of MORB and also on ocean island basalts over a wide range of MgO contents. The $^{88}$Sr value of MORB is 0.31 ± 0.05‰, which is close to those of the bulk silicate earth (0.30 ± 0.02‰) and carbonaceous chondrites (0.29 ± 0.06‰) (Amsellem et al., 2018). Regarding different host-rock types, basalts, granites, and andesites have similar average values (Moynier et al., 2010; Charlier et al., 2012; Amsellem et al., 2018). The calculated end-member fluids of the sediment-starved sites (0.284 ± 0.030‰) suggest that $^{88}$Sr is essentially independent of rock type (basalt, dacite) and hydrothermal setting (volcanic front, back-arc basin). Therefore, the $^{88}$Sr values of end-
members of group B exhibit minimal impact on hydrothermal sites, which suggests that the phase separation and segregation from host rocks. Ishibashi et al. (1994b) and Yamaoka et al. (2015) and have similar North Fiji Basin sites are all hosted by MORB-like basalt (Ishibashi et al., 1994b) and have similar physicochemical conditions (Table 2) with their $\delta^{88}\text{Sr}$ values being in the range of oceanic basalt (Amsellem et al., 2018).

The LHOS field in the North Fiji Basin had a slightly lower $\delta^{88}\text{Sr}$ end-member value (0.26‰) compared with those of the other phase-separated sites. We wish to address the possibility that it is lower due to the impact of anhydrite precipitation/re-dissolution, as well as hydrothermal calcite veins. Voigt et al. (2018) conducted an experiment, demonstrating that the formation of hydrothermal anhydrite and the preferential dissolution of $^{86}\text{Sr}$-enriched minerals occurs during seawater-basalt reactions at 250°C and 290°C, together, these processes account for 0.07–0.18‰ decrease in the $\delta^{88}\text{Sr}$ values of fluids after a few months of reaction time between seawater and basaltic materials. Because $\text{Sr}^{2+}$ co-precipitation in anhydrite serves as the main $\text{Sr}$ sink in hydrothermal reactions, we used $\text{Ca}^{2+}$ and $\text{SO}_4^{2-}$ concentrations to infer the contributions of rock dissolution and anhydrite precipitation (Figure 7A) to the end-member $\delta^{88}\text{Sr}$ values. The vectors in Figure 7A show the directions associated with anhydrite precipitation (a decrease in the 1:1 M ratio) and rock dissolution (predominantly the release of $\text{Ca}^{2+}$ on the scatter diagram of $\text{Ca}^{2+}$ and $\text{SO}_4^{2-}$. The overall magnitude of the rock dissolution effect relative to the anhydrite precipitation effect was less at phase-separated sites than those of the other sediment-starved sites (Figure 7A). An impact of anhydrite precipitation on the lower $\delta^{88}\text{Sr}$ values of LHOS site was also evaluated using a simple closed/semi-closed type fractionation (Supplementary Figure S2). The anhydrite-fluid fractionation factor $\varepsilon_{\text{anhydrite-fluid}}$ of 0.034 ± 0.011‰ (Voigt et al., 2018) was used for calculation. We considered two extreme cases: 1) anhydrite precipitation from recharged seawater (0.392‰), and 2) anhydrite precipitation from end-member fluids of other sediment-starved sites in the North Fiji Basin (0.285‰). In the former case, 98% removal of $\text{Sr}$ by anhydrite is necessary to obtain the LHOS end-member of 0.256‰; in the latter case, 58% is necessary. However, $\text{Sr}$ concentrations of the LHOS end-member (66 μmol/kg) is about half of that of seawater (89 μmol/kg), and higher than other end-members in the North Fiji Basin (36 μmol/kg for White Lady, 56 μmol/kg for Kaiyo, Table 2). Because of the similarity of the end-member $\delta^{88}\text{Sr}$ values among the sediment-starved sites, $\text{Sr}$ co-precipitation with anhydrite cannot by itself account for the lower $\delta^{88}\text{Sr}$ end-member value at the LHOS site. Therefore, the variation in $\delta^{88}\text{Sr}$ value of oceanic basalt (0.31 ± 0.05‰, Amsellem et al., 2018) would be an explanation for the lower $\delta^{88}\text{Sr}$ value of LHOS site. Similar to anhydrite, the impact of $\text{Sr}$ co-precipitation within hydrothermal calcite veins is also considered to be negligible, as calculated from the magnitude of $\text{Ca}$ depletion observed in the hydrothermal fluids and the partitioning coefficient for $\text{Sr}$ to $\text{Ca}$ ratios (Rimstidt et al., 1998).

The difference between the flux-weighted marine $\delta^{88}\text{Sr}$/$\delta^{87}\text{Sr}$ input and the intercept value has been attributed to an elevated supply of riverine $\text{Sr}$ to the ocean due to post-glacial weathering (Figure 6; Krabbenhöft et al., 2010; Pearce et al., 2015a). Our data imply that the mean marine $\delta^{88}\text{Sr}$ composition of hydrothermal input may be closer to an average oceanic crustal value of approximately 0.30‰ (Amsellem et al., 2018), as it gives a higher intercept $\delta^{88}\text{Sr}$ value (Figure 6).

### Influence of Host Sediments on Okinawa Trough Hydrothermal Fluids

The $\text{Sr}$ isotopes in the end-member hydrothermal fluids from the sediment-hosted vents in the Okinawa Trough have $^{87}\text{Sr}/^{86}\text{Sr}$ values distinctly higher than the other vent (approximately 0.710–0.709 vs. < 0.705, Noguchi et al., 2009) as well as distinctly lower $\delta^{88}\text{Sr}$ values (approximately 0.22 vs. 0.28‰). The higher $^{87}\text{Sr}/^{86}\text{Sr}$ values most likely reflect hydrothermal interaction with the clastic fraction of the host sediments.
Despite the similar sedimentary origins of D621-5, D622-1 and D622-2, two vents at the Minami-Ensei site, the chemical data differ considerably between them (Table 1). The chemical characteristics of the D622-2 vent fluid, including the end member Sr concentration of 252 µM, are comparable to those of the vent fluids of Minami-Ensei Knoll reported by Kawaguchi et al. (2013). Although D621-5 is chemically similar to the other sediment-hosted samples, D622-2 exhibited higher Sr/Ca ratios and 87Sr/86Sr values that were trend outliers. Previous studies have reported that the fluid/sediment mass ratio during interactions of circulating hydrothermal fluids with terrigenous sediments determines the leaching of fixed and adsorbed trace elements from sediments (Yamaoka et al., 2013; Araoka et al., 2016). Therefore, inter-vent variability is also related to heterogeneity of the sediments (such as relative abundance of hemipelagic sediments to volcanic deposits) and the scales of subseaﬂoor hydrothermal fluid flow structures at sediment-hosted sites.

CONCLUSION

We investigated δ88Sr in submarine vent ﬂuids from arc and back-arc hydrothermal systems in the western Paciﬁc. The important observation of this study is that the end-member δ88Sr values (0.256–0.298‰, calculated by extrapolating the linear regression line ﬁtted to data from two or three samples from each site to a Mg concentration of zero) are similar to the average value of oceanic volcanic rocks, about 0.28–0.31‰ (Amsellem et al., 2018). This result conﬁrms the utility of the δ88Sr mass balance for past changes in the marine carbonate system (Krabbenhöft et al., 2010; Vollstaedt et al., 2014; Pearce et al., 2015a), and supports the view that hydrothermal end-member in sediment-starved hydrothermal systems is probably a unique δ88Sr value through geologic time because of the similarity and homogeneity of stable Sr isotopes in between igneous rocks as well as carbonaceous chondrites (Amsellem et al., 2018). The decreased δ88Sr values, as low as 0.20%, accompanied by higher 87Sr/86Sr, in vent ﬂuids of the sediment-hosted sites are linked to the release of Sr from sedimentary materials, that we ﬁnd to be sensitive to the extent of mixing between the different sources and sinks of sedimentary Sr phases. A global average of hydrothermal δ88Sr value would be reassessed with taking into account these data and other representative hydrothermal system, such as the East Paciﬁc Rise.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

TY, SW, and HK contributed to conception of the study. Samples were collected by TG. TY, SW, and TI performed the isotopic
analysis. DA performed the trace metal analysis. TY wrote the first draft of the manuscript with input from NO, TI, and SW. SW wrote the analytical sections of the manuscript. All authors contributed to manuscript revision, read, and approved the submitted version.

**FUNDING**

This work was undertaken with the support of Japan Society for the Promotion of Science (JSPS) to TY (no. 16H05883, 19K21908), to SW (no. 18K03814) and to TG (no. 16H02701).

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**ACKNOWLEDGMENTS**

We thank Kazuya Nagaishi and Tatsuya Kawai of Marine Work Japan for their support during the analysis. The suggestions of Thomas M. Blattmann greatly improved the language. The comments of the reviewers are gratefully acknowledged.

**SUPPLEMENTAL MATERIAL**

The Supplemental Material for this article can be found online at: [https://www.frontiersin.org/articles/10.3389/feart.2020.591711/full/supplementary-material](https://www.frontiersin.org/articles/10.3389/feart.2020.591711/full/supplementary-material)
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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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