Supporting Information

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Possible Sources of Hydrothermal Vent Input to the Study Locations

It is impossible to identify the exact vent sources for the hydrothermally derived dFe observed at the three study locations (Fig. 2). It is also probable, given that the study sites are situated far away from a large array of known hydrothermal vents, that multiple single vents contribute to the cumulative hydrothermal dFe and ^{3}He anomalies measured at the three stations. We review here all of the possible hydrothermal vent fields that may have contributed to the observed dFe enrichment, in light of the recently assembled global hydrothermal vent database (1) as well as estimates of abyssal circulation pathways.

Southwest Pacific: KM0703 SPEEDO Station 19 (20°S, 170°W). The hydrothermal dFe, dMn, and excess ^{3}He data at Station 19 are elevated over a depth range from 1,000 to 3,500 m, with coincident maxima at 2,000 m. Although the nearest identified hydrothermal sources occur in the Tonga–Fiji region just 600–700 km to the west of this station (2), the predominant flow pattern over this depth range is from the east, organized as a zonally elongated anticyclonic gyre extending as far eastward as the East Pacific Rise, according to Reid (3). This suggests that the fast-spreading centers (centered between 2,000 and 3,000 m depth) along the southern East Pacific Rise (EPR) described by Lupton and Craig (4) are the most likely origin of the ^{3}He and dFe anomalies 6,000 km away at KM0703 SPEEDO. Moreover, the 2,000 m depth circulation at Station 19 from the southeast is consistent with the beta-plume dynamics earlier described by Stommel (5), which arise from the middepth injection of buoyancy at these sites. We discount hydrothermal contributions from the Lau Basin because it is downstream of KM0703 Station 19. Although a ^{3}He plume has been observed north and west of Station 19 (2), centered at 1,750 m near 15°S, 175°W, it emanates westward toward the Coral Sea with tight ^{3}He contours connoting a sharp drop-off of ^{3}He concentrations between 173° and 176°W, supporting the hypothesis of little eastward influence of Lau Basin vents on the dFe and ^{3}He profiles at Station 19 (170°W). Furthermore, none of the stations studied by Lupton et al. (2) along 170°W show evidence of the ^{3}He signal coming from the Lau Basin; these stations also show very little ^{3}He variation with latitude, which would be expected if it originated from the Lau Basin. Using all of these lines of evidence, we are very confident in our conclusion that the KM0703 Station 19 signal derives from the EPR ~6,000 km to the east.

Southeast Pacific: Melville BiG RAPA Stations 4 (23.5°S, 88.75°W) and 7 (26.25°S, 104°W). The hydrothermal ^{3}He and dFe anomalies at Stations 4 and 7 in the southeast Pacific both reach maxima at 2,000 m depth, with Station 7 having >50% more dFe than Station 4 at their maxima, although the ^{3}He anomalies are roughly comparable in magnitude at the two stations. The highly elevated dFe concentrations of >1.5 nmol/kg at 2,000 m at Station 7 might be interpreted as indicative of a proximal Fe source. This station is also located on a topographic high on the Easter Seamount Chain, which might also point to a nearby hydrothermal source. However, no venting has been confirmed or even inferred anywhere along the Easter Seamount Chain (as shown by a lack of hydrothermal vent crosses along this zonal topographic rise in Fig. 1). The Sala y Gomez hot spot, which formed the Eastern Seamount Chain over the last 30 million years (6, 7), is currently believed to be somewhere between Easter Island (540 km away from Station 7) and Sala y Gomez Island (145 km away), likely nearer to Sala y Gomez Island. No modern seamount derived from this hot spot has been identified, and as a result no volcanic activity or associated venting has been inferred.

Thus, the EPR is the nearest possible source of dFe and ^{3}He at Melville BiG RAPA Station 7. The well-known ^{3}He plume initially discovered at ~15°S between 2,000 and 3,000 m is advected westward from the EPR, away from the Peru/Chile basins (4), but there is still elevated δ^3He (28–30%) at 15–30°S in the Peru Basin east of the EPR through and along 88°W (8) (the longitude of Station 4). Despite being east of the EPR, Lupton (8) attributed this 88°W δ^3He distribution to an EPR source because steric height calculations infer eastward transport of water south of 20°S at 2,500 m depth into the Peru/Chile basins (9). In fact, the ^{3}He distribution along 32°S very clearly shows eastward transport of a ^{3}He plume from the southern EPR (10). This eastward aybssal flow south of 20°S has also been used to explain the enhanced Fe and Mn in surface sediments of the Peru/Chile Basins at 25°S (11) and the enhanced total dissolved Mn concentrations (from unfiltered samples) collected at 18–25°S, 94°W near Station 4 of our study (12). As shown in Fig. 1, there are many hydrothermal vents between 20 and 32°S (13) that could source this enhanced dFe, dMn, and ^{3}He along the Nazca Ridge, most of which vent at the appropriate depths of 2,000–3,000 m to produce the observed signal. These EPR vents are a minimum of 860 km from Station 7 and thus indicate that dFe can be stably transported quite far from the vent source.

The geostrophic flow patterns discussed by Reid (3) at Station 4 flow from the west–northwest at 2,000–2,500 m depth, instead of from due west, as at Station 7. Thus, although it is most likely that Station 4 dFe is derived from the southern EPR due to its closer venting source, it may also receive some hydrothermal influence from the northern EPR. As a hydrothermal plume extends away from its vent site, it typically shoals along isopycnal surfaces, and consequently, ^{3}He plumes near 32°S, 80°W in the Chile Basin were hypothesized by Jenkins (10) to have originated in the North Pacific and then shoaled during transport to the South Pacific because the South Pacific enhanced ^{3}He was at a shallower isopycnal than southern EPR-derived vent plumes. It is not clear whether a hydrothermal dFe signal from the North Pacific would survive the long transit times to the South Pacific, but it is possible that North Pacific vents may contribute a portion of the accumulated hydrothermal signal observed in the southeast Pacific. Station 4 is at a minimum of 2,400 km from southern EPR vents, and hydrothermal signals would have had to transit even farther if derived from northern EPR vents.

dMn Data and Fe/Mn Ratios

Hydrothermally-derived dissolved Mn (dMn) maxima were detected at 2,000 m in our open ocean samples at all three stations, ranging from 0.32 to 1.18 nmol/kg in the profile peaks (compared with 0.1–0.2 nmol/kg dMn backgrounds in typical abyssal seawater). Station 7 in the eastern South Pacific had the highest dMn concentration, coincident with the highest dFe concentrations. Station 4 (23.5°S, 88.75°W) had a peak dMn concentration of 0.40 nmol/kg, which is much lower than the 1.77 nmol/kg total dissolved Mn (dissolved + particulate) measured in 1986 at a nearby station (24.64°S, 94.04°W) at 2,579 m depth (12). Assuming similar hydrothermal Mn delivery at the two sampling stations today and in 1986, there must be a large particulate Mn component at these stations that was not monitored in this study, potentially suggesting that both particulate and dissolved Mn are transported

hundreds to thousands of kilometers from the vent source in the hydrothermal plume. This particulate Mn inference warrants further investigation for confirmation.

Because of its slow scavenging kinetics (14) and sluggish equilibration following temperature alteration in the buyoant plume (15), dMn is believed to behave conservatively in vent fluids on the timescale of hours to days. Following this assumption of Mn conservation, dFe/dMn ratios have been used in vent fluid samples to calculate the extent of plume dilution (16, 17) and the temperature of the hydrothermal reaction chamber (18). Because our samples are taken from the distal hydrothermal plume at a very minimum of several days from the vent source, we believe that an analysis of vent fluid dilution using the dissolved Mn data is not appropriate because dissolved Mn no longer behaves conservatively on these timescales, and ^{3}He is instead used as the conservative tracer of vent fluids in this paper.

However, there is still useful information to be gleaned from the Fe/Mn ratios with distance from the plume, although background deep ocean concentrations must be subtracted from the hydrothermal concentration anomalies to compare the distal hydrothermal Fe/Mn ratios to near-field hydrothermal ratios. We made these corrections by subtracting the nonhydrothermal, deep ocean dFe and dMn concentrations of 0.4 nmol/kg and 0.15 nmol/kg, respectively (19). The resulting dFe/dMn ratios at the depths of maximum hydrothermal influence are as follows: Station 19, 2.65 ± 0.5; Station 7, 1.16 ± 0.2; and Station 4, 2.21 ± 0.6. Assuming that both Fe and Mn behave nonconservatively in the far-field plume, these ratios inform us about the relative scavenging precipitation potential of Fe and Mn in the distal hydrothermal plume. If we assume the same hydrothermal source for Stations 4 and 7, with Station 4 simply ~1,500 km farther downstream in the distal plume, then the increase in dFe/dMn ratio from 1.16 at Station 7 to 2.21 at Station 4 suggests that dMn is scavenged to a greater extent in the distal plume than dFe, presumably because more of the scavengeable dFe has already been removed in the near-field plume. This is in contrast to a study by Hawkes et al. (20) of hydrothermal vents along the East Scotian Ridge, where proximal to the vent site, dMn appeared to be depleted from the dissolved phase faster than dFe. Although dFe removal pathways along the distal plume are abiotically driven, including oxidation of remaining dissolved Fe^{2+} or pyrite nanoparticles and aggregation/scavenging of colloidal Fe to the particulate phase, dMn removal pathways include both an abiotic component (oxidation of dissolved Mn^{2+} to particulate Mn^{4+}) as well as a biotic component that is microbially mediated (21). This microbial Mn scavenging was the dominant Mn removal pathway in distal (15–20 km from vent) samples, compared with a larger abiotic scavenging component in proximal vent samples (0–3 km from vent) near the Juan de Fuca Ridge (14).

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Other Supporting Information Files

Table S1 (XLSX)