Basin-scale transport of hydrothermal dissolved metals across the South Pacific Ocean

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1   **Hydrothermal venting along mid-ocean ridges exerts an important control on the chemical**

2   **composition of seawater by serving as a major source or sink for a number of trace**

3   **elements in the ocean[1–3]. Among these, iron has received considerable attention because of**

4   **its role as an essential and often limiting nutrient for primary production in regions of the**

5   **ocean that are of critical significance for the global carbon cycle[4]. It was thought that most**

6   **of the dissolved iron discharged by hydrothermal vents is lost from solution close to ridge-**

7   **axis sources[2,5] and of limited importance for ocean biogeochemistry[6]. This long-standing**

8   **view is challenged by recent studies that suggest that stabilization of hydrothermal**

9   **dissolved iron may facilitate its long-range oceanic transport[7–10]. Such transport has been**

10   **subsequently inferred from spatially limited oceanographic observations[11–13]. Here we**

11   **report data from the U.S. GEOTRACES Eastern Pacific Zonal Transect (EPZT) that**

12   **unequivocally demonstrate lateral transport of hydrothermal dissolved iron, manganese,**

13   **and aluminium from the southern East Pacific Rise (SEPR) more than 4000 km westward**

14   **across the South Pacific Ocean. Dissolved iron exhibits unexpected conservative behaviour**

15   **in this hydrothermal plume, implying a greater longevity in the deep ocean than previously**

16   **assumed[6,14]. Based on our observations, we estimate a global hydrothermal dissolved iron**

17   **input of 3-4 Gmol y^{-1} to the ocean interior, which is more than four-fold higher than**

18   **previous estimates[7,11,14]. Complementary simulations with a global-scale ocean**

19   **biogeochemical model suggest that the observed transport of hydrothermal dissolved iron**

20   **requires some means of physicochemical stabilization and indicate that hydrothermally**

21   **derived iron sustains a significant fraction of Southern Ocean export production.**

22

23   Hydrothermal fluids are enriched in iron (Fe) and manganese (Mn) by more than 10^6 relative

24   to ambient deep ocean concentrations[1], and corresponding gross hydrothermal fluxes to the

25   oceans are likely greater than those from global riverine inputs[3]. However, it has been well-

26   documented that most of the hydrothermal Fe is lost from the dissolved phase in the vicinity of

27   ridge-axis vents, where hot (~350°C), acidic, anoxic hydrothermal fluids ascend and mix with

28   cold, alkaline, oxic seawater, resulting in the formation of Fe-sulphides and/or Fe-

29   oxyhydroxides[2,5], which are subsequently lost from solution due to settling and scavenging. As a

30   result of these removal processes, it has been widely assumed that seafloor hydrothermal

31   emissions are not a major source of dissolved Fe (DFe) to the ocean[6]. In contrast, dissolved Mn

32   (DMn) is oxidized more slowly than DFe in seawater, and hydrothermal DMn anomalies have

33   been observed as far as 2000 km from ridge-axis sources[15].

34   A number of recent studies have demonstrated that DFe can be stabilized against

35   precipitation, aggregation, and scavenging losses from seawater by several different physico-

36   chemical mechanisms[7–10]. Such findings imply that hydrothermal activity could exert an

37   important influence on the oceanic DFe inventory; however, comprehensive observational data

38   on the persistence and fate of hydrothermal Fe are needed to evaluate this hypothesis. Although

39   several recent studies have inferred the transport of hydrothermal DFe over distances of

40   hundreds[16,17] to thousands of kilometers[11–13], those conclusions remain equivocal, owing to

41   limited sampling coverage[11–13] and assumptions regarding corresponding distributions of the

42   hydrothermal tracer helium-3 (^{3}He)[11,13,16].

43   Here we present data for samples collected from 35 hydrographic stations between Manta,

44   Ecuador, and Papeete, Tahiti, during the Eastern Pacific Zonal Transect (EPZT; U.S.

45   GEOTRACES cruise GP16; Fig. 1). This expedition focused attention on the Peru upwelling

46 region and the superfast-spreading southern East Pacific Rise (SEPR), one of the most

47 volcanically active areas on Earth and the source of a well-documented plume of hydrothermal

48 ^{3}He that extends west across the deep South Pacific Ocean[18]. The data from this cruise reveal

49 pronounced gradients in DFe, DMn, dissolved aluminium (DAl), and excess-^{3}He (^{3}He$_{xs}$)

50 concentrations along the ~8000 km cruise transect (Fig. 2). The most striking feature, and the

51 main focus of this communication, is a vast, mid-depth plume of elevated DFe and DMn that

52 extends over an unprecedented distance of more than 4000 km to the west of the SEPR. This

53 plume is carried by the westward flowing mid-depth circulation[18–20], and is clearly defined by

54 anomalous concentrations of the hydrothermal tracer ^{3}He$_{xs}$ (Fig. 2). The distance over which DFe

55 and DMn are transported from the SEPR is substantially greater than that observed in plumes

56 identified from basin-scale sections across the Atlantic, Indian, Arctic, and Southern

57 oceans[12,16,17,21–23]. Also notable are the elevated DAl concentrations that extend more than 3000

58 km west of the SEPR; DAl enrichments of this magnitude and extent have not been previously

59 reported for hydrothermal plumes in the Pacific, Arctic, Southern, or Indian oceans[12,21,22,24,25]

60 (see also: *GEOTRACES Intermediate Data Product*, 2014). Given the differences in

61 geochemical behaviour between Fe and Mn, it is surprising that the lateral extent of the

62 hydrothermal DFe anomaly exceeds that of DMn: inventories of hydrothermal DFe and DMn

63 (DFe and DMn minus background) at station 32 are ~11% and ~4%, respectively, of those at

64 station 20 (see Fig. 1). Our data set unequivocally documents the long-range transport of

65 hydrothermal DFe from the SEPR, thus confirming the tentative conclusions drawn from limited

66 previous observations[11,13].

67 Directly over the SEPR at Station 18, DFe in the 'near field' hydrothermal plume is only ~20%

68 of total dissolvable Fe (an approximate measure of total hydrothermal Fe), and Fe(II)

69   concentrations are near background. This suggests rapid oxidation and loss of hydrothermal Fe

70   from the dissolved phase close to the ridge axis, consistent with previous observations from the

71   SEPR[5]. In contrast, from the first off-axis station (Station 20) continuing west across the basin as

72   far as Station 36, DFe concentrations are linearly correlated with ^{3}He$_{xs}$ within the plume (Fig. 3a,

73   b and Extended Data Fig. 1), indicating nearly conservative dilution of the hydrothermal DFe

74   over a distance of ~4300 km. Such behaviour is completely unexpected, given the known

75   propensity for the oxidation, aggregation, and scavenging of DFe from seawater[2,4,5].

76   Accordingly, our observations imply that hydrothermal DFe in the plume is somehow stabilized

77   against loss from solution, perhaps as a result of complexation by dissolved organic ligands[7,8], or

78   by incorporation in inorganic or organic nanoparticles that reside within the dissolved (<0.2 μm)

79   size fraction[9,10].

80   The relationship between DMn and ^{3}He$_{xs}$ in the plume (Fig. 3c, d) indicates that hydrothermal

81   Mn is removed from the dissolved phase until it reaches Station 21, beyond which the residual

82   hydrothermal DMn, like DFe, behaves conservatively with respect to ^{3}He$_{xs}$. DAl over the ridge

83   crest is enriched by as much as 12 nM over mid-depth concentrations to the east of the SEPR.

84   This is comparable to enrichments in hydrothermal plumes over the Mid-Atlantic Ridge [23,26],

85   where Al-rich plumes are spatially restricted to the deep axial valley and are thought to reflect

86   the entrainment of Al-rich waters by rising hydrothermal fluids during plume formation[26]. In

87   contrast, the SEPR has no significant axial valley, and the DAl plume extends far from the ridge

88   crest, suggesting a more significant source of DAl along the SEPR. DAl concentrations >100

89   nM have been reported in unusually acidic hydrothermal plumes that may be associated with

90   seafloor eruptive activity[27], and the SEPR between 14°S and 19°S is a particularly active locus

91   of seafloor volcanism, with hydrothermal and eruptive activity being more intense than along

92   most other ridge sections worldwide[5,28]. This suggests eruptive activity is one possible source of

93   the SEPR DAl plume.

94   We assess the importance of physicochemical stabilization to the long-range transport of

95   hydrothermal DFe using numerical simulations of DFe and $^3He_{xs}$ within a global-scale ocean

96   biogeochemical model that includes explicit cycling of DFe-binding ligands (see Methods). The

97   model represents the input of ^3He as a function of ridge spreading rate and simulates

98   hydrothermal Fe efflux via a fixed Fe:^3He ratio based on a global compilation of hydrothermal

99   fluids[14]. Although there is reasonable qualitative agreement with our $^3He_{xs}$ data (Extended Data

100   Fig. 2), the sluggish deep ocean circulation typical of relatively coarse-resolution global models

101   restricts the overall westward transport of the hydrothermal anomaly (a feature that has proven a

102   challenge to measure and model[19,20]). Even allowing for this insufficient abyssal propagation,

103   DFe concentrations and their relation to $^3He_{exs}$ decrease rapidly to the west of the ridge crest due

104   to scavenging when only hydrothermal Fe input is considered (Fig. 4a, b and Extended Data Fig.

105   3a); a tenfold increase in Fe input results in no improvement in the agreement with observations

106   (Fig. 4a, b and Extended Data Fig. 3b). In contrast, when DFe-stabilizing 'ligands' from vent

107   fluids or from processes occurring within the plume are added in an equimolar ratio with

108   hydrothermal DFe, significant westward propagation of the DFe plume is achieved and the

109   model is better able to reproduce both the plume extent and relationship between DFe and $^3He_{xs}$

110   (Fig. 4a, b and Extended Data Fig. 3c-e). Adding tenfold more ligands, or tenfold more ligands

111   and tenfold more hydrothermal Fe increases the plume extent still farther (note logarithmic scale

112   in Fig. 4a,b). Importantly, including a hydrothermal supply of ligands also improves the degree

113   to which the model can reproduce the global distributions of DFe in the abyssal ocean (Extended

114   Data Table 1). A similar result would be expected for the formation of relatively unreactive Fe

115 nanoparticles within the near-field plume[9,10], thus the DFe-stabilizing 'ligands' could involve

116 organic or inorganic moieties. These process-based model experiments indicate that the total

117 input of hydrothermal Fe regulates the magnitude of the DFe plume near the ridge crest, whereas

118 the stabilization of DFe against loss from solution governs its persistence and transport in the

119 deep ocean.

120   Although recent global-scale models of the ocean Fe cycle suggest significant hydrothermal

121 contributions to the deep-ocean DFe inventory[14], our data indicate that this previous work

122 substantially underestimated the far-field influence of hydrothermal DFe emissions. The linear

123 relationship between DFe and $^3He_{xs}$ concentrations in the plume west of the SEPR has a slope of

124 $7.5 \pm 0.8 \times 10^6$ mol mol^{-1} (s.d. of slope based on simple linear regression; Fig. 3b),, which falls

125 roughly midway between values estimated for hydrothermal plumes in the western South Pacific,

126 the Southern Ocean, and the South Atlantic[11,14,16]. If this relationship is representative of steady-

127 state mid-ocean ridge hydrothermal inputs to the ocean, then the estimated global hydrothermal

128 ^3He efflux of 530 mol y^{-1} [29] yields an 'effective' hydrothermal DFe input of ~4 ± 1 Gmol y^{-1} to

129 the ocean interior, which is at least fourfold higher than previous estimates[7,11,14]. This

130 hydrothermal DFe is ultimately supplied to the iron-deficient surface waters of the Southern

131 Ocean, where it supports ~15-30% of the modelled export production south of the Polar Front

132 (Fig. 4c). The impact of hydrothermal Fe on export production is driven by both its gross flux

133 and processes that govern its stabilization (Extended Data Fig. 4). Thus, the ultimate impacts of

134 hydrothermal activity on the biogeochemical cycle of Fe in the ocean may depend as much on

135 the processes that control the longevity of hydrothermal DFe plumes as on the magnitude of the

136 hydrothermal DFe emissions, on which prior studies have largely focused.

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207

219 **Author Contributions:** JAR participated on the EPZT and determined DAl and DMn; PNS

220 interpreted the DFe data; CRG co-designed the study and participated in the EPZT; WJJ

221 collected [3]He data; JWM co-designed the study, participated in the EPZT, and collected

222 Fe(II) data; BMS participated in the EPZT and determined DFe; AT conducted the

223 modelling experiments and interpreted their results. All authors contributed to the writing

224 of the manuscript.

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227 **The authors declare no competing financial interests.**

228   **Figure 1. Cruise track and station locations.** The U.S. GEOTRACES Eastern Pacific Zonal

229   Transect cruise (GEOTRACES GP16) was undertaken on R/V *Thomas G. Thompson* cruise 303

230   from 25 October to 20 December 2013. Station locations are shown as yellow circles with station

231   numbers in white. Station 18 is located over the crest of the East Pacific Rise.

232   **Figure 2. Interpolated zonal concentration for GEOTRACES Eastern Pacific Zonal**

233   **Transect cruise. a,** Dissolved iron (DFe). **b**, Dissolved manganese (DMn). **c**, Dissolved

234   aluminium (DAl). **d**, Excess helium-3 (^{3}He$_{xs}$). Station numbers and distance west of East Pacific

235   Rise are indicated on uppermost panel.

236   **Figure 3. Relationship between dissolved trace metals and ^{3}He west of SEPR. a,** Dissolved

237   iron (DFe) *vs* excess helium-3 (^{3}He$_{xs}$) at 2500m depth. **b**, DFe *vs* ^{3}He$_{xs}$ integrated over 2200-

238   2800 m depth except at Station 18, where the maximum depth was 2640 m. **c**, Dissolved

239   manganese (DMn) *vs* ^{3}He$_{xs}$ at 2500 m depth. **d**, DMn *vs* ^{3}He$_{xs}$ integrated as in b. Error bars are 2

240     $\times$ the relative standard deviation of a given analysis as reported in the Methods. Error bars are

241   absent where the symbol size exceeds the error estimate. Lines represent the slope of a simple

242   linear regression analysis of the data. Discrete and integrated ^{3}He$_{xs}$ concentrations are lower at

243   Station 18 relative to stations west of the ridge; this difference is reduced for integrations

244   between 2200 and 2640 m depth (Extended Data Figure 1a). The relatively low ^{3}He$_{xs}$

245   concentrations at Station 18 (~15°S) suggest that the off-axis plume (Stations 20-36) is primarily

246   derived from vent fields located farther south (~17°S-18.5°S) along the SEPR[5,28] with

247   hydrothermal and eruptive effluent being homogenized and transported north and west[20] by

248   along- and off-axis transport processes[20,30].

249   **Figure 4. Results of biogeochemical model simulations.** Model simulation results centred at

250   2530 m depth (average of two depth bins spanning 2060–3010 m; coloured lines) are shown in

251   panels a and b. **a,** DFe from model results compared to measured DFe concentrations (crosses)

252   between 2200 and 2800 m to the west of the ridge axis. **b**, DFe versus excess helium-3 from

253   model simulations compared to measured values (diamonds) at 2500 m depth. For panels **a** and **b**

254   the individual model simulations were run for 75 years and are identified as follows: Orange

255   dashed line is model solution using base hydrothermal Fe flux ($1 \times$ Fe); Red dashed line is for

256    $10 \times$ base hydrothermal Fe flux ($10 \times$ Fe); Cyan line is base hydrothermal Fe flux with equimolar

257   flux of ligands ($1 \times$ Fe + $1 \times$ ligands); Dark blue line is for base hydrothermal Fe flux with $10 \times$

258   greater ligand flux ($1 \times$ Fe + $10 \times$ ligands); Green line is for $10 \times$ base Fe flux with equimolar

259   flux of ligands ($10 \times$ Fe + $10 \times$ ligands). **c**, Percent of annual export production due to

260   hydrothermal Fe based on a 500-year model simulation employing base hydrothermal Fe flux

261   with equimolar ligand flux ($1 \times$ Fe + $1 \times$ ligands) relative to a model solution with no

262   hydrothermal Fe or ligand flux. Lower export production in the subtropical oceans is caused by

263   decreased preformed macronutrients in the mode waters.

264

METHODS

Sample collection and processing. Water column samples for trace metal analyses were obtained using 24 modified 12 L GO-FLO bottles (General Oceanics) mounted on a trace-metal clean conductivity-temperature-depth (CTD) carousel (SeaBird) that was deployed on a Kevlar conducting cable [31]. Upon recovery, the GO-FLO samplers were brought into a shipboard Class-100 clean laboratory container for sub-sampling. For filtered samples, the samplers were pressurized to 10 psi using filtered, compressed air, and the seawater samples were filtered through pre-cleaned 0.2 µm Acropak Supor capsule filters (Pall) using rigorous trace-metal clean protocols[31]. The Eastern Pacific Zonal Transect (EPZT) occupied 35 sampling stations along more than 8000 km of cruise track. At 17 stations denoted as 'full' or 'super' stations, 37 samples were collected between the surface and the seafloor; at one additional station (Station 34), 25 samples were collected over the upper 3000 m of the water column. At 13 stations denoted 'demi' stations, 13 samples were collected in the upper 1000 m; and at 4 stations over the continental shelf, 7-24 samples were collected between the surface and seafloor. For dissolved manganese and aluminum (DMn and DAl), 876 samples were collected from all stations and depths. The 0.2 µm filtered subsamples were stored in 100 mL low density polyethylene (LDPE) bottles (Bel-Art) with LDPE caps and were acidified to pH ~1.7 with 12 N ultrapure hydrochloric acid (Fisher Optima). For dissolved iron (DFe), 760 samples were collected at the full and super stations, with 0.2 µm filtered subsamples stored in 125 mL LDPE bottles with polypropylene caps (Nalgene) and acidified to pH ~1.7 with a 6 N solution of ultrapure hydrochloric acid (Fisher Optima). Unfiltered seawater samples for the analysis of total-dissolvable iron and manganese (TDFe and TDMn) were collected at all full stations west of 109°W from the 24 deepest samples. These samples were collected in 125 mL LDPE bottles

288   with polypropylene caps (Nalgene) and acidified to pH ~1.7 with 12 N ultrapure hydrochloric

289   acid (Fisher Optima), then stored for > 4 months prior to analysis. Sample collection for iron(II)

290   (Fe(II)) analysis was identical to that for total dissolved metals, with the exception that the

291   seawater samples were drawn into acid-washed 50 mL AirTite™ All-Plastic Norm-Ject™

292   Syringes (Fisher Scientific) in the GEOTRACES sampling van to exclude oxygen

293   contamination. These samples were stored on ice and in darkness to slow oxidation prior to

294   analysis. Comparison with samples where Fe(II) was stabilized using 3-(N-

295   morpholino)propansulfonic acid buffer [32] indicated that Fe(II) was effectively preserved using the

296   syringe protocol. Independent measurements of DFe indicated no detectable contamination from

297   the syringes. Seawater samples for dissolved helium analysis (~45 g each) were drawn from the

298   standard rosette (12-position, 30 L Niskin-type bottles) using Tygon tubing connected to lengths

299   of 5/8″ soft copper refrigeration tubing. Sample tubes were then hydraulically crimp-sealed[33].

300   **Analytical methods.** DFe was determined at sea or at Old Dominion University by flow

301   injection analysis with in-line pre-concentration on resin-immobilized 8-hydroxyquinoline and

302   colorimetric detection[34,35], using a method modified from *Measures et al.* [36]. For the lowest

303   concentration samples from each analysis, and the SAFe S reference seawater, the method of

304   standard additions was used; all other samples were quantified using a standard curve obtained

305   by addition of Fe standard solution to low-iron seawater. For the cruise period, we determined

306   the following DFe concentrations for the SAFe seawater reference materials: 0.126 ± 0.023 nM

307   (n = 4) for SAFe seawater reference material S, and 1.26 ± 0.20 nM (n = 10) for SAFe seawater

308   reference material D2. These values compare well with community consensus concentrations of

309   0.095 ± 0.008 nM and 0.955 ± 0.024 nM, respectively. In an effort to correct for day-to-day

310   variations in analytical accuracy, all daily analyses included the GEOTRACES GSP seawater

311 reference material, for which there is currently no consensus DFe concentration; all daily sample

312 determinations were corrected using the difference between each day's measured GSP

313 concentration and the overall cruise average DFe concentration for the GSP seawater (0.34 ±

314 0.07, n = 27). The analytical limit of detection is estimated as the DFe concentration equivalent

315 to a peak area that is 3 times the standard deviation on the 'zero-loading blank' ('manifold

316 blank'), from which we estimate a detection limit of less than 0.04 nM[34,37]. Blank contributions

317 from the ammonium acetate sample buffer solution (added on-line during analysis) and

318 hydrochloric acid (added after collection) are negligible (i.e., too low to quantify). Robust

319 estimates of analytical precision are derived from multiple separate determinations of the SAFe

320 seawater reference materials, which yield analytical uncertainties (expressed as one relative

321 sample standard deviation on the mean) of ~15% at the concentration level of SAFe S (~0.1 nM)

322 and ~10% at the concentration level of SAFe D2 (~1 nM). For high Fe (> 5 nM) samples, TDFe

323 and DFe were determined by modifying the flow injection method to include a sample loop,

324 rather than a pre-concentration column, and by using deionized water acidified to pH ~1.7 as a

325 carrier in place of the acid eluent. This modified flow injection method had an analytical

326 precision of ± 4% or ± 1.5 nM (whichever is greater). Suitable seawater reference materials

327 were not available for these analyses.

328 DMn was determined at sea by flow injection analysis with in-line pre-concentration on resin-

329 immobilized 8-hydroxyquinoline and colorimetric detection[38]. Daily precision of analysis was ±

330 0.01 nM (one standard deviation) or 3.8%, whichever is larger, based on the reproducibility of

331 analytical and internal standards. A conservative estimate of the limit of detection is 0.03 nM

332 based on 3 times the daily precision of analysis, which is consistent with previous work[38]. Two

333 internal reference standards were run over the 57 days of the cruise, with DMn concentrations of

334 0.42 ± 0.036 nM (± 8.4%; n = 102) and 0.31 ± 0.041 nM (± 13%, n = 69), respectively. The

335 SAFe reference samples were analyzed simultaneously during sample analysis with the

336 following results: for SAFe S, 0.85 ± 0.026 nM (n = 27; consensus value = 0.79 ± 0.06 nM); for

337 SAFe D2, 0.40 ± 0.028 nM (n = 22; consensus value = 0.35 ± 0.05); and for SAFe D1, 0.36 ±

338 0.026 nM (n = 31; no consensus value). Analytical Uncertainty is expressed as ± one standard

339 deviation.

340 DAl was determined at sea by flow injection analysis with in-line pre-concentration and

341 fluorimetric detection[39]. Method modifications included replacing resin-immobilized 8-

342 hydroxyquinoline with Toyopearl AF-Chelate 650M, and using acidified de-ionized water as the

343 carrier instead of acidified seawater. Daily precision for repeat analysis of internal and primary

344 standards was ± 0.1 nM or 4.2%, whichever is larger. Two internal reference standards were run

345 during the cruise, with DAl concentrations of 1.76 ± 0.25 nM (± 14%; n = 101) and 1.98 ± 0.07

346 nM (± 3.4%; n = 75), respectively. The SAFe reference samples were analyzed simultaneously

347 during sample analysis; for SAFe S, 2.38 ± 0.14 nM (n = 26; consensus value = 1.74 ± 0.09 nM);

348 for SAFe D2, 1.63 ± 0.13 nM (n = 26; consensus value 1.04 ± 0.1 nM); and for SAFe D1, 1.26 ±

349 0.11 nM (n = 32; consensus value = 0.64 nM). The least squares best fit between our shipboard

350 determinations (DAl$_s$) and the SAFe consensus values is (DAl$_c$) is: DAl$_s$ = 1.02 DAl$_c$ + 0.59 nM

351 (r^2 = 0.99). Analytical Uncertainty is expressed as ± one standard deviation. In the past, our

352 laboratory has produced DAl determinations that were statistically indistinguishable from the

353 SAFe consensus concentrations, suggesting that our shipboard analytical method includes a

354 consistent, unidentified blank equivalent to ~0.6 nM DAl. Our estimated limit of detection of

355 0.3 nM based on daily precision estimates is low and might more conservatively be estimated to

356 be > 0.6 nM. The anomalously high DAl concentrations (3.7–29.5 nM) determined in samples

357   collected from 20-150 m depth at stations near 109°W and 113°W are not readily explained by

358   ancillary chemical and physical data from the cruise, although there is no apparent reason to

359   suspect that these few samples were contaminated during collection, processing, or analysis.

360   Fe(II) was determined at sea using an automated flow injection analysis system (FeLume II,

361   Waterville Analytical) employing a luminol chemiluminescence detection system [32,40]. The

362   FeLume system was fitted with a standard quartz flow cell and a Hamamatsu HC135 photon

363   counter configured with the following settings: pump speed of 15 revolutions per minute; photon

364   counter integration time of 200 ms; load time of 20 s. The mean of the last 50 data points was

365   used to determine the signal. Detection limits were determined for surface samples where

366   ferrous iron was negligible based on a standard 3σ evaluation of the baseline signal [32,40]. This

367   yielded an estimated detection limit of 14 pmol L^{-1}.

368   Helium was determined ashore after gases from the samples were quantitatively extracted under

369   a vacuum into liquid-nitrogen chilled ~25 mL aluminosilicate glass flasks and sealed prior to

370   analysis. Sample processing on the mass spectrometer system included purification over SAES

371   getters to remove reactive gases and use of cryogenics to separate the noble gases[41–43]. Sample

372   integrity was evaluated using noble gas abundances (not reported here), and determined by

373   quadrupole mass spectrometer with an accuracy of 0.1-0.5%, depending on the gas. The helium

374   abundance and isotope ratio (^{3}He/^{4}He) were determined using a branch-tube magnetic sector

375   mass spectrometer to an accuracy of 0.15% or better as determined by reproducibility of

376   standards and duplicate samples. The isotope ratio was referenced to an atmospheric standard.

377   Excess ^{3}He (^{3}He$_{xs}$) is computed as an approximate measure of the non-atmospheric ^{3}He over

378   saturation:

379 $$^3He_{xs} = (\delta^3He - \delta*^3He)/100 \times C[He] \times 1.384 \times 10^{-6}$$

380 Where $\delta^3He = 100 \times (R_x/R_a - 1) \times 100\%$, R_x and R_a are the $^3He/^4He$ ratios of the sample and air

381 (1.384×10^{-6}), respectively. $\delta*^3He$ is the helium isotope ratio anomaly in solubility equilibrium

382 with the atmosphere, which is a weak function of temperature[44] and is $\approx$ -1.8% for the data used

383 here. The precision of $^3He_{xs}$ is 0.5% measurements was 1 fM. The precision of $^3He_{xs}$ is more than

384 tenfold better than that of either DFe or DMn allowing the use of a Type I linear regression when

385 comparing DFe and DMn to $^3He_{xs}$.

386 **Details of the PISCES biogeochemical model.**

387 The model employed in this study is currently the only global-scale version that considers

388 hydrothermal input of iron and a dynamic representation of iron-binding ligands[45]. The PISCES

389 model[46,47] is a relatively complex ocean general circulation and biogeochemistry model that

390 includes two phytoplankton groups, two zooplankton grazers, five limiting nutrients (nitrate,

391 phosphate, silicic acid, ammonium, and Fe) and two size classes of organic carbon particles,

392 calcium carbonate and biogenic silica, which sink and are remineralized differentially. DFe is

393 supplied to the ocean from dust, sediments, rivers, and hydrothermal vents[47] and is ultimately

394 lost to the sediments. The DFe is subjected to scavenging/coagulation losses, which produce two

395 size classes of particulate iron. The scavenging rate is computed in the model by calculating the

396 amount of 'free' uncomplexed DFe (assuming a dynamic ligand concentration and conditional

397 stability) and the resulting net rate of scavenging depends on the concentrations of each particle

398 species. We also account for the loss of organically-complexed colloidal iron via coagulation

399 processes and consider contributions from turbulent and Brownian components. The colloidal

400 fraction of DFe is calculated as a function of temperature, salinity, and pH[48,49]. Ligand dynamics

401   are represented assuming sources from phytoplankton or zooplankton exudation and organic

402   matter degradation, and sinks associated with photochemical degradation, colloidal coagulation,

403   and variable bacterial consumption, all on a reactivity continuum[45]. The ligand stability constants

404   vary according to: $pKFe'L = 17.27 - 1565.7/TK$ where TK is absolute temperature leading to

405    $pKFe'L$ of 11.5 at 0 °C and 11.9 at 20 °C[45]. Phytoplankton DFe uptake is computed using a quota

406   model, with overall growth limitation accounting for the iron demand associated with

407   photosynthesis, respiration, and nitrate uptake[50]. PISCES is coupled to the three dimensional

408   ocean general circulation model NEMO, which has a spatial resolution of 2° of longitude, $2 \times$

409   cos(latitude) that is enhanced to 0.5° at the equator, and 31 vertical levels, with the first 10 levels

410   in the upper 100 m. Hydrothermal transport was mostly observed over vertical levels 25

411   (centered 2290 m, depth range 2050 to 2530 m) and 26 (centered at 2770 m, depth range 2530 to

412   3010 m), which were used in Figs. 4a and 4b.

413   For this study we conducted a range of different simulations with PISCES aimed at addressing

414   the processes responsible for the longevity of the observed hydrothermal DFe plume and their

415   potential impact on the carbon cycle. The standard input flux of iron from the mid ocean ridge

416   was calculated based on iron to helium-3 ratios in hydrothermal fluids and spreading rate[14]; note

417   the flux is not based on the *dissolved* Fe flux calculated here. First, we conducted a set of

418   experimental simulations over 75 years outlined in Fig. 4 to examine the plume extent. To assess

419   the large-scale impact of hydrothermal Fe and 'hydrothermal ligands' on ocean biogeochemistry

420   and productivity, we extended the run with the standard addition of hydrothermal DFe with

421   ligands in a 1:1 molar ratio ($1 \times Fe + 1 \times$ ligands) over a period of 500 years and compared that

422   to a 500-year model run in which no hydrothermal Fe or ligands were added. After 500 years, the

423   yearly change in biogeochemical tracers was negligible. In the model, ligands decay with time

424 (microbial decay). As a result, the addition of hydrothermal ligands does not lead to their

425 unrealistic accumulation in the ocean, with the main anomaly decaying rapidly from the ridge

426 crest (Extended Data Fig. 5). Overall, the addition of hydrothermal ligands in the '1 $\times$ Fe + 1 $\times$

427 ligands' and '1 $\times$ Fe + 10 $\times$ ligands' experiments increases the total ligand inventory from 1.18 $\times$

428 10^9 to 1.35 $\times$ 10^9 mol. To isolate the effect of hydrothermal ligand supply, we conducted a

429 simulation that added only hydrothermal ligands without hydrothermal DFe and compared it to

430 an experiment in which no ligands were added (Extended Data Fig. 5). This experiment is likely

431 not representative of the real ocean, because ligands that might be produced at or near

432 hydrothermal vents would be saturated with the DFe supplied by hydrothermal vents. As a

433 consequence this experiment releases ligands into the ocean with an extremely high capacity to

434 complex DFe from other sources. In this extreme hypothetical case where unsaturated

435 hydrothermal ligands are able to bind DFe from other sources, only a small (~5%) increase in

436 export production in the Southern Ocean is observed.

437 We have conducted a statistical analysis of the model against the most recent compilation of DFe

438 averaged onto the World Ocean Atlas grid. We note that it is challenging to quantitatively

439 evaluate global-scale iron models as we are obliged to compare to localized point measurements

440 rather than having an objective climatology such as those available for macronutrients (e.g.,

441 World Ocean Atlas). Both the model and the observations were gridded onto 1° $\times$ 1° grid with 33

442 vertical levels. After log transforming over 2000-5500 m depth, there are 1025 unique data

443 comparisons. In the abyssal ocean (2000-5500 m depth) we find support for the conclusions

444 drawn from the visual model-data comparison in Fig. 4. The correlation (Extended Data Table

445 1) increases markedly when a source of hydrothermal ligands is applied, relative to the model

446 runs that do not add hydrothermal ligands (1 $\times$ Fe and 10 $\times$ Fe).

447   **Hydrothermal plume inventory estimates.** Depth-integrated metal inventories for depth

448   intervals of interest were estimated by summing the product of the average concentration of

449   samples from two sequential depths and the difference in depth between those samples. Where

450   duplicate samples were collected to provide overlap between hydrocast sampling, the average

451   concentration of the samples from overlapping depths was used.

452   **Ocean Data View Parameters and Adjustments (Figure 2).** Excess helium-3 concentration

453   data were contoured using Diva gridding with a signal-to-noise of 10. Dissolved iron

454   concentration data were contoured using Diva gridding with a signal-to-noise of 6.5 with

455   negative values suppressed. Dissolved manganese concentration data were contoured using Diva

456   gridding with a signal-to-noise of 4 with negative gridded values suppressed. Dissolved

457   aluminum concentration data were contoured using Diva gridding with a signal-to-noise of 11

458   with negative gridded values suppressed.

459   The extremely high concentrations of these species over the ridge crest (Station 18) resulted in

460   interpolated concentrations at Station 17 that vastly exceeded the actual measured

461   concentrations. To circumvent this contouring artefact, an artificial background station was

462   inserted at 111.5°W, which is halfway between the ridge-crest station (Station 18) and the first

463   station to the east (Station 17). This 'background station' duplicated the measured depth and

464   concentration data for each species from Station 17. The black sample location indicators for

465   this artificial station were removed from Fig. 2.

466   **Cruise track selection.** The latitude for the western portion of the EPZT cruise was selected to

467   follow the 'downstream' core of the hydrothermal helium-3 (^{3}He) plume close to latitude 15°S, as

468   determined from observations of previous research expeditions in the eastern South Pacific

469   Ocean (GEOSECS, HELIOS, and WOCE programs). Urabe *et al.*[28] surveyed the SEPR axis

470   between 13.8°S and 18.6°S, finding the most intense hydrothermal plumes between 17°S and

471   18.5°S. These plumes were rich in particulate iron[5] and total dissolvable manganese (TDMn)[51].

472   The TDMn concentrations at both 2500 m depth and integrated over 2200-2800 m depth in the

473   most intense plumes over the SEPR axis in 1993 were greater than corresponding values

474   observed at any of our EPZT cruise stations. As the ^{3}He data are unpublished, we examine

475   ^{3}He:TDMn along the ridge crest [28], which has the highest values between 17°S and 18.5°S,

476   suggesting that both discreet and integrated ^{3}He concentrations were much higher in those

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527

Figure Captions for Extended Data

Extended Data Figure 1. Depth integrated concentrations of **a** dissolved Fe (DFe), and **b** dissolved Mn (DMn), versus depth integrated concentration of excess helium-3 ($^3He_{xs}$), over a depth range of 2200-2640 m. Sample station numbers are indicated for each data symbol.

Extended Data Figure 2. Comparison of modeled (rectangles) and measured (circular symbols) concentrations of excess helium-3 ($^3He_{xs}$), between EPZT cruise Station 36 (far left) and Station 17 (far right).

Extended Data Figure 3. Sections of modeled DFe transport and decay using a dynamic ligand global-circulation model[45] (see Methods). The model scenarios listed here are the same as those presented in Figure 4. **a,** 1 x Fe; **b**, 10 x Fe; **c**, 1 x Fe + 1 x ligands; **d**, 1 x Fe + 10 x ligands; **e**, 10 x Fe + 10 x ligands.

Extended Data Figure 4. Impacts on carbon export from model simulations. **a**, Percentage contribution to carbon export production due to the input of hydrothermal DFe, not considering the addition of hydrothermal ligands. **b**, Additional percentage contribution from the addition of hydrothermal ligands to the simulation shown in panel a. This panel represents the difference between the total impact from the addition of both hydrothermal DFe and ligands (manuscript Figure 4c) compared to the input hydrothermal DFe without the addition of ligands shown in panel **a**, above.

Figure 5. Ligand Flux Model Experiments. Two experiments were run to assess the impact of the flux of ligands associated with hydrothermal activity on the oceanic budget. a. Model

548   simulation with no hydrothermal ligand flux. b. Model simulation with ligand flux equal to the

549   flux of hydrothermal Fe.

South Pacific Ocean

Manta
Rinse 2
Peru
Rinse 3

35 33
36   34   32
         30   28   26
29   27   25
23
24   22
21 18
20
17
16
15
14
13
12
11
10   8   6
9   7
4 2
5 3

Papeete

160°W   140°W   120°W   100°W   80°W

0
10°S
20°S

a

4,300 km

depth (m): 0, 1,000, 2,000, 3,000, 4,000, 5,000

DFe (nM): >5.0, 2.5, 2.0, 1.5, 1.0, 0.5, 0

Station numbers: 36, 34, 32, 30, 28, 26, 25, 23, 21, 20, 18, 17, 15, 13, 11, 9, 7, 1

b

DMn (nM): >2.0, 1.5, 1.0, 0.5, 0

0.2, 0.4

c

DAl (nM): >12, 10, 8, 6, 4, 2, 0

4, 6

d

^{3}He$_{xs}$ (fM): 1.4, 1.2, 1.0, 0.8, 0.6, 0.4

longitude: 140° W, 120° W, 100° W, 80° W

Ocean Data View / DIVA

Concentration at 2,500m **Integrated from 2,200 to 2,800m**

a

$Y = 6.4 \times 10^6 X - 5$
$R^2 = 0.96$

DFe (nM)

b

$Y = 7.5 \times 10^6 X - 3500$
$R^2 = 0.94$

DFe (μmol/m^2)

c

$y = 1.9 \times 10^6 X - 2$
$R^2 = 0.97$

DMn (nM)

d

$Y = 2.5 \times 10^6 X - 1190$
$R^2 = 0.86$

DMn (μmol/m^2)

^{3}He$_{xs}$ (nM x 10^{-6})

^{3}He$_{xs}$ (μmol/m^2 x 10^{-6})

a

more Fe supplied
greater Fe stabilization

westward Transport

log DFe (nM)

150°W   140°W   130°W   120°W   ridge crest

longitude

b

10 x Fe + 10 x ligands
1 x Fe + 10 x ligands
1 x Fe + 1x ligands
10 x Fe
1 x Fe

$^3He_{xs}$ (fM)

c

latitude

longitude

% impact on carbon export