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# 1 Pathways and timescales of Southern Ocean hydrothermal iron and 2 manganese transport

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13

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15 revisions. AJB, CAB, NJW, KP, HJV, CMM, IT, AM, SJU and APM participated in the field campaign. AJB  
16 and NJW conducted trace metal (micronutrient) analysis. KP performed macronutrient analysis. HJV  
17 processed conductivity-temperature-depth sensor package datasets. CAB performed Lagrangian  
18 modelling experiments and analysis. SO carried out the water mass analysis. APM, CMM, SJU and AM  
19 wrote and planned the original research questions and funding proposal. APM led the research  
20 program. The views expressed in this paper are those of the authors alone, and not the organisations for  
21 which they work.

22 **Abstract**

23 Scarcity of iron (Fe) and manganese (Mn) limits the efficiency of the biological carbon pump over large  
24 areas of the Southern Ocean (SO). The importance of hydrothermal vents as a source of these  
25 micronutrients to the euphotic zone of the SO is debated. We present full depth profiles of dissolved  
26 and total dissolvable trace metals in the remote eastern Pacific sector of the SO (55-60°S, 89.1°W),  
27 providing evidence of enrichment of Fe and Mn at depths of 2000-4000 m. These enhanced  
28 micronutrient concentrations were co-located with <sup>3</sup>He enrichment, an indicator of hydrothermal fluid  
29 originating from ocean ridges. Modelled water trajectories revealed the understudied South East Pacific  
30 Rise and the Pacific Antarctic Ridge as likely source regions. Additionally, the trajectories demonstrate  
31 pathways for these SO hydrothermal ridge-derived trace metals to reach the SO surface mixed layer  
32 within two decades, potentially supporting a regular supply of micronutrients to fuel SO primary  
33 production.

## 34 1.0 Introduction

35 Despite being the largest high-nutrient low-chlorophyll region<sup>1,2</sup>, the Southern Ocean (SO) is a  
36 globally important organic carbon sink, with the interplay between circulation and remineralisation  
37 dynamics setting the rate of carbon uptake by the biological carbon pump<sup>3</sup> and 10% of global biological  
38 carbon export occurring in the region<sup>4</sup>. Iron (Fe) is an essential but often scarce micronutrient that can  
39 limit primary production and the efficiency of the biological carbon pump in the SO<sup>5,6</sup>. However, other  
40 elements can potentially be (co-)limiting alongside Fe in the Southern Ocean (SO)<sup>7</sup>, with recent evidence  
41 for manganese (Mn) (co-)limitation in particular<sup>2,8-10</sup>.

42 As non-conservative elements with residence times on the order of decades in the deep  
43 ocean<sup>11</sup>, the distributions of dissolved Fe and Mn (dFe, dMn; <0.2  $\mu\text{m}$ ) in seawater are spatially coupled  
44 to sources. The main external sources of Fe and Mn to the ocean are margin sediments, atmospheric  
45 dust, and hydrothermal venting<sup>6,12</sup>. Input from margin sediments enhances primary production  
46 downstream of islands in the SO<sup>13,14</sup>, and sustains ecosystems in Antarctic shelf regions<sup>12,15</sup>, where  
47 melting glacial ice can be an additional source of Fe and Mn<sup>16,17</sup>. Marine aerosol Fe concentrations can  
48 vary by >3 orders of magnitude, causing sporadic and seasonal changes to the widespread deposition of  
49 Fe to the surface ocean<sup>18</sup>. However, according to both observations and models, the remote southern  
50 hemisphere oceanic gyres and polar regions have some of the lowest Fe aerosol deposition fluxes in the  
51 world (<0.01 g Fe m<sup>-2</sup> yr<sup>-1</sup>)<sup>19</sup>.

52 Both Fe and Mn are concentrated in hydrothermal fluids, often enriched by a factor of >10<sup>6</sup>  
53 relative to background seawater<sup>20</sup>. Early studies suggested that Fe and Mn entering the ocean from  
54 hydrothermal vents was precipitated or scavenged and sedimented close to vent sites<sup>12,21</sup>, with  
55 precipitation reported to occur before the plume reaches neutral buoyancy<sup>22</sup>. However, it is now known  
56 that a small, but significant, fraction of the dFe supplied from hydrothermal vents is sufficiently  
57 stabilised against precipitation to be transported in the ocean interior<sup>23-25</sup>. Hydrothermal venting is

58 estimated to supply  $4 \pm 1$  Gmoles dFe yr<sup>-1</sup> to the wider deep ocean away from the proximal vent  
59 sites<sup>23,26</sup>, representing a continuous Fe input in contrast to short term variations observed in other Fe  
60 sources, such as atmospheric deposition<sup>26</sup>. Similarly, observed increases in deep ocean dMn  
61 concentrations have been attributed to hydrothermal activity<sup>16</sup>, and incorporating hydrothermal dMn  
62 inputs increases the accuracy of modelled oceanic dMn distributions<sup>12</sup>. Our understanding of the  
63 hydrothermal plume processes responsible for the physico-chemical stabilisation of Fe is developing,  
64 though not yet comprehensive. Within hydrothermal plumes dFe has been shown to form inorganic  
65 nanoparticles<sup>27,28</sup>, larger inorganic colloids<sup>29</sup> and organic complexes<sup>30,31</sup>. The co-location of carbon with  
66 Fe in plume particles suggests that organic carbon may alter the chemical behaviour of Fe  
67 oxyhydroxides<sup>29</sup> and create localised regions of Fe(II) enrichment<sup>31</sup>.

68         Due to the inhibition of vertical mixing in the ocean interior by density stratification,  
69 hydrothermal plumes tend to travel predominantly along isopycnals<sup>32,33</sup>. Westerly winds drive the  
70 Antarctic Circumpolar Current (ACC) to create a northward surface flow (Ekman Transport), causing  
71 isopycnals to shoal in the SO<sup>34</sup>. Consequently, upwelling deep waters in the SO may provide a pathway  
72 for hydrothermal trace metals to be mixed into the SO euphotic zone<sup>21,35</sup>. Inputs of hydrothermal dFe  
73 from shallow (<500m) and deep (>2000m) vents have been linked to regional phytoplankton blooms  
74 within<sup>36,37</sup> and outside of the SO<sup>38</sup>. The potential for hydrothermal dFe to sustain primary production in  
75 the SO has been evaluated in models<sup>39,40</sup>, but consensus on the significance of this source of Fe and Mn  
76 has yet to be reached.

77         To sustain SO primary production, upwelling timescales must be short enough to deliver  
78 hydrothermal Fe and Mn to the euphotic zone before removal processes, such as scavenging and  
79 precipitation, deplete these metals. Assuming that some degree of physicochemical stabilisation of dFe  
80 in neutrally buoyant plumes may allow transportation to surface waters, model results indicate that  
81 hydrothermal Fe could support  $\approx 15$ -30% of export production south of the Polar Front<sup>39,41</sup>. However,

82 estimates of dFe residence time in the East Pacific Rise (EPR) far field plume at 15°S have been revised  
83 from quasi-conservative, based on comparisons with <sup>3</sup>He measurements<sup>23</sup>, to non-conservative with a  
84 residence time of 9-50 years, based on comparisons with <sup>228</sup>Ra measurements<sup>42</sup>. The 9-50 year residence  
85 time estimate is consistent with a global modelling study estimate of hydrothermal Fe residence time of  
86 21-35 years<sup>39</sup>. Similarly, deep ocean dMn residence times are estimated at 5-40 years<sup>11</sup>.

87           In comparison, transit time estimates for the shoaling of Circumpolar Deep Waters (CDW),  
88 originating in the Pacific (≥30°S), to the SO mixed layer range from 17 years to a few centuries<sup>43-46</sup>. Such  
89 uncertainty surrounding hydrothermal Fe input and stabilisation processes, alongside SO ventilation  
90 rates, is reflected in Fe modelling studies. A global steady state inverse circulation model combined with  
91 a mechanistic Fe model was used to conclude that only 3-5% of hydrothermal dFe reaches surface  
92 waters globally<sup>40</sup>. A subsequent study argued however that the inverse modelling approach reduces the  
93 magnitude of hydrothermal Fe inputs from ridge systems located within the SO relative to a spreading  
94 rate model approach<sup>39</sup>. This ongoing debate is hindered by a lack of observational evidence of the key  
95 transport pathways. The Southeast Pacific sector of the SO has been indicated as a critical region for  
96 hydrothermal Fe and Mn input by models<sup>12,39</sup>, but is a 'data desert' for trace metal observations. In this  
97 study, we provide the first full depth profiles of Fe and Mn from this SO region, which show a clear  
98 midwater hydrothermal trace metal signal. By modelling water pathways, we trace the hydrothermal  
99 signal back to specific ridge systems and evaluate whether the observed hydrothermal Fe and Mn could  
100 supply the SO mixed layer, and therefore fuel SO primary production.

## 101 2.0 Results and Discussion

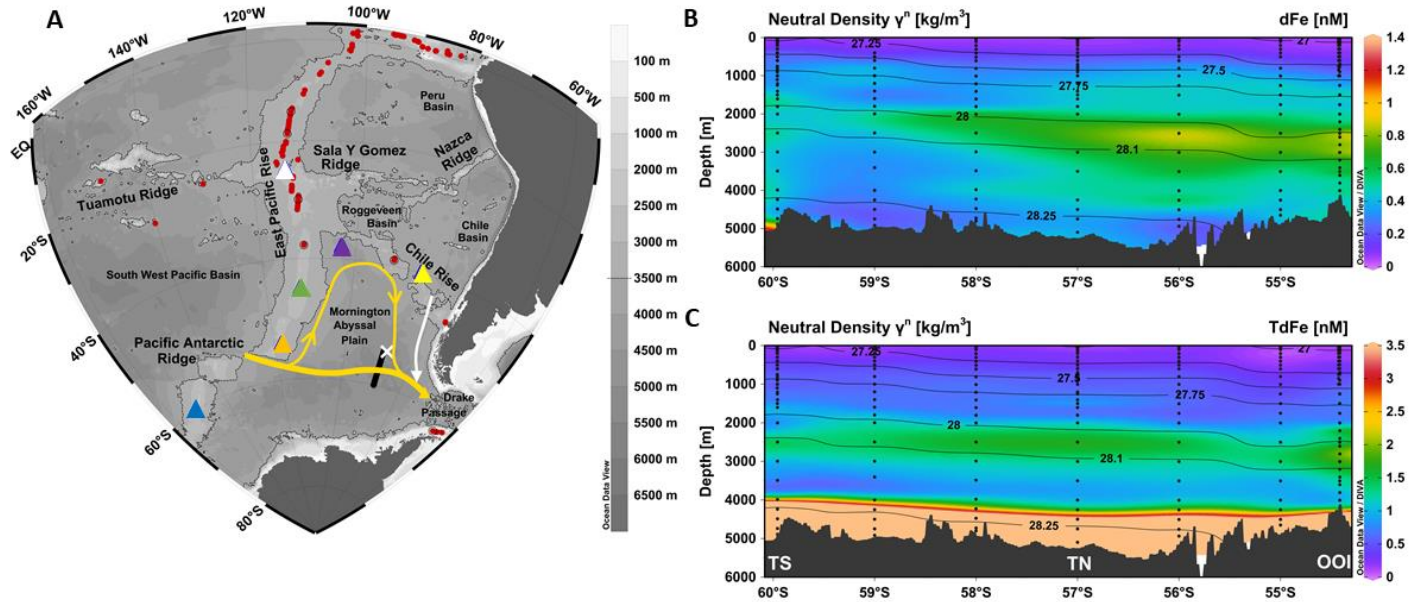
### 102 2.1 Observations of hydrothermal trace metals in the southeast Pacific sector of the Southern 103 Ocean

104 Our study sampled a north-south transect along 89°W located in the Morington Abyssal Plain  
105 (MAP) in the southeast Pacific Ocean (Fig. 1 A). As expected, SO surface waters were characterised by  
106 extremely low dFe (<0.2 µM, 0-30 m depth;  $0.09 \pm 0.04$  nM,  $n= 18$ ) and total dissolvable Fe (TdFe;  
107 unfiltered) concentrations (0-30 m depth;  $0.19 \pm 0.09$  nM,  $n= 13$ ; Fig. 1B, C) and excess macronutrient  
108 concentrations (nitrate + nitrite 16.6-23.6 µM, phosphate 1.0-1.6 µM; data not shown), consistent with  
109 previous upper ocean observations in this region<sup>47</sup> and wider SO biogeochemistry<sup>35</sup>.

110 The most striking feature of our transect was Fe enrichment in deep waters (Fig. 1B, C), with dFe  
111 concentrations of 0.48 to 0.96 nM occurring between 2000-4000 m at the northern end of our transect  
112 (Ocean Observatories Initiative (OOI) sustained observatory location, 54.08°S 89.67°W  
113 <https://oceanobservatories.org/array/global-southern-ocean-array/> ; Fig. 1A, Table S1). Higher TdFe  
114 concentrations (>2.5 nM with a maximum concentration of 15.4 nM), associated with lower dFe  
115 concentrations, were also observed in benthic nepheloid layers driven by particle accumulation and  
116 benthic sediment resuspension of particulate Fe and scavenging of dFe (Fig. 1C, Fig 2D.). For  
117 comparison, high particulate Fe concentrations (up to 88 nM) were observed above the seafloor at open  
118 ocean stations in the north Atlantic<sup>48</sup>.

119

120



121

122 **Figure 1. A** Study area in the southeast Pacific and Southern Ocean. Red dots are the location of known

123 active vent sites<sup>49</sup>. Lack of exploration means that knowledge of active vent site locations over the

124 region is incomplete. The yellow arrow is the approximate flow of water at 2000-4000 m depth<sup>50</sup> and

125 white arrow the approximate flow of Southeast Pacific Deep Slope Water<sup>51</sup>. The solid black line is the

126 transect occupied by this study. The white cross is the sampling location of a previous <sup>3</sup>He profile<sup>52</sup>

127 collected in 1993 approximately 83 km northeast (bearing of 055°) of our northernmost station.

128 Triangles indicate the start locations for forward-tracked trajectory modelling in this study: yellow - East

129 Chile Rise, purple – West Chile Rise, white – Easter Microplate, green – North East Pacific Rise, orange –

130 South East Pacific Rise (SEPR) and blue – Pacific Antarctic Ridge (PAR). **B** and **C** are section plots of dFe

131 (<0.2 μm) and TdFe (unfiltered) concentrations along the transect with neutral density overlain. Black

132 dots mark sampling depths. Note that concentrations in **B** and **C** are on different scales and that TdFe

133 concentrations were typically >2.5 nM near the seafloor. The maximum concentration measured was

134 15.39 ± 1.12 nM. OOI is the sampling location coinciding with the Ocean Observatories Initiative

135 mooring. Stations OOI, TN and TS (panel C) are the locations of trace metal depth profiles displayed in

136 Figure 2.

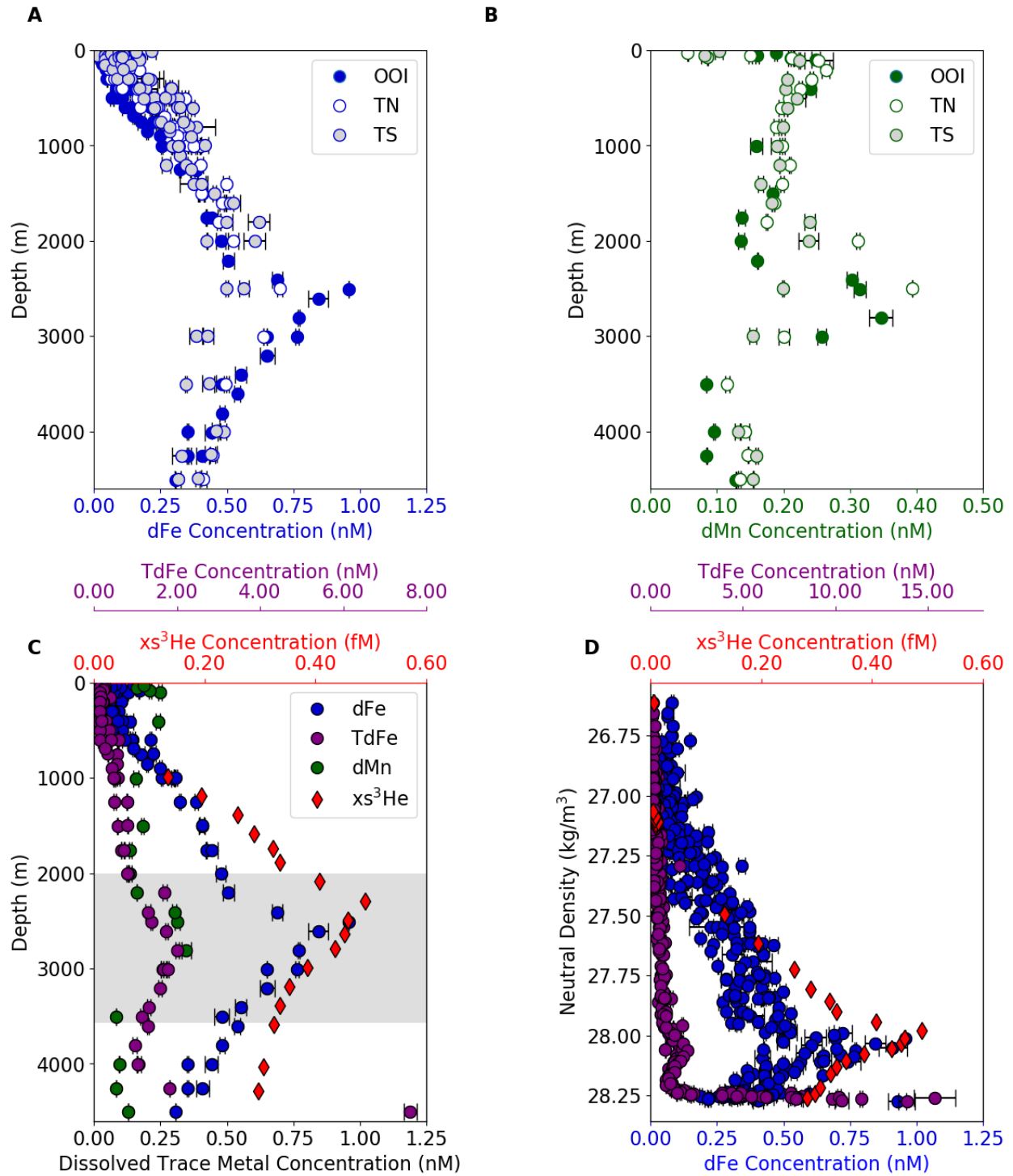


137           Away from external sources, deep ocean dMn concentrations are typically very low (<0.2 nM)  
138 due to scavenging removal into the particulate phase and oxidation of Mn ions to insoluble Mn oxides<sup>16</sup>.  
139 However, like dFe, basin-scale transport in vent plumes has been observed e.g. westward transport of  
140 dMn from the EPR (10-17°S) occurs in the low latitude Pacific Ocean<sup>23</sup>. Along our transect we present,  
141 elevated dMn concentrations (up to 0.39 nM, Table S1) occurred coincidentally with the enriched dFe  
142 signal ( Fig. 2A, B and C), compared to a background dMn concentration (~0.2 nM) immediately above  
143 and below the mid-depth enrichment ( Fig. 2B and C). In the southwest Pacific (170°W), CDW has  
144 background dMn concentrations of ~0.1-0.2 nM<sup>53</sup> away from hydrothermal and sediment inputs. The Fe  
145 and Mn enrichment we observed was centered on a neutral density surface of  $\gamma_n = 28.0-28.1 \text{ kg m}^{-3}$  (Fig.  
146 2D), within CDW (Fig. S1). This density surface shoals by ~880 m towards the south of the transect as  
147 part of the wider Ekman driven upwelling occurring in the SO. The influence of this wind-driven  
148 upwelling is shown clearly in the maximum dFe concentrations, which tracked the isopycnals along the  
149 depth section (Fig. 1) and in the dMn profiles from stations OOI, TN and TS (Fig. 1B, C and Fig.2A and B,  
150 Table S1).

151           Mantle helium (He), which is enriched in primordial <sup>3</sup>He, is also released into the ocean at mid-  
152 ocean ridges. A ratio of <sup>3</sup>He/<sup>4</sup>He in excess of atmospheric values (hereafter  $x_s^3\text{He}$ ) in deep ocean waters  
153 is a widely used tracer of hydrothermal inputs<sup>52</sup>. Tagliabue et al. (2022) indicated that the basin-scale  
154 hydrothermal Fe supply from SO vents is not well predicted by the spatial distribution of <sup>3</sup>He inputs,  
155 likely because of inter-vent Fe:<sup>3</sup>He ratio variability. Nevertheless, at a single location the coincident  
156 enrichment of <sup>3</sup>He, Fe and Mn in deep ocean waters is indicative of hydrothermal input of these  
157 elements. Helium isotope observations, previously sampled near station OOI (Fig. 1), revealed the  
158 presence of  $x_s^3\text{He}$  at the depths and neutral density of our observations of trace metal enrichment (Fig.  
159 2 C,D). Although the helium dataset 26 years, and 83 km away from our trace metal observations, the  
160 comparable distribution of dFe, TdFe and  $x_s^3\text{He}$  with neutral density (Fig. 2D) provides confidence that

161 we observed a consistent oceanographic signal in both datasets. However, we cannot account for any  
162 temporal variability of end-member metal and helium concentrations during the 26-year gap between  
163 trace metal and helium sample collection<sup>54</sup>. The Fe concentration maximum at the northern end of our  
164 study site was slightly deeper (215 m) than the mantle  $x\text{s}^3\text{He}$  maximum. A similar offset in the  
165 distribution has been observed further north in the far field plume of the EPR (20-26°S) and has been  
166 attributed to reversible scavenging of dFe onto sinking particles, which deepens the metal concentration  
167 maximum<sup>55</sup>. The offset we observed may also be due to temporal variability of end member metal and  
168 helium concentrations, the influence of multiple vent sites with varying Fe: $^3\text{He}$  ratios, and/or reversible  
169 scavenging combined with layering of fine particulate material with irregular element distribution within  
170 plumes<sup>41</sup>.

171         The balance between the release of dFe from remineralising organic matter and removal of dFe  
172 from solution via scavenging processes leads to an accumulation of dFe in Pacific Mode and  
173 Intermediate Waters<sup>56</sup>. We show that the signal we observed is decoupled from the mineralization of  
174 biogenic particles by plotting dFe against apparent oxygen utilisation (AOU; Fig. S2). In waters below the  
175 mixed layer there is a positive linear relationship between dFe and AOU indicating that remineralisation  
176 of sinking organic matter exerts an important control on dFe concentrations, consistent with previous  
177 investigations of Pacific mode and intermediate waters<sup>56</sup>. However, the enrichment of dFe we observe  
178 in deeper waters clearly deviates from the linear dFe-AOU trend indicating an additional deep water dFe  
179 source such as long range transport of hydrothermal dFe<sup>24</sup>



180

181 **Figure 2. A** Depth profiles of dFe at stations OOI, TN and TS. **B** Depth profiles of dMn at stations OOI, TN

182 and TS. **C** Depth profiles of dissolved trace metals (dFe and dMn) and TdFe at our northernmost station

183 OOI (combined Fe data from 4 site visits) and nearby historic  $xs^3He$  (see Fig. 1A; white cross). **B** All dFe  
184 and TdFe data from the transect and nearby historic  $xs^3He$  plotted against neutral density.

## 185 **2.2 Identifying the source region of the observed hydrothermal signal**

186 The bathymetry of the southeast Pacific Ocean has several mid-ocean ridge systems (Fig. 1A),  
187 the most prominent being the EPR which is aligned meridionally at  $\sim 115^\circ W$  at depths of around 2500 m.  
188 The southern extension of the EPR, the Pacific Antarctic Ridge (PAR), is aligned zonally ( $\sim 55-65^\circ S$ ). The  
189 Chile Rise ( $\sim 40-45^\circ S$ ) bounds the North of the Morington Abyssal Plain (MAP; our study region)  
190 between the EPR and the coast of Chile. The OOI site is a little south of the confluence of the easterly  
191 flowing ACC and the eastern boundary current. The latter flows within  $\sim 1500$  km of the coast of South  
192 America between 1500 m and 3500 m, this Southeast Pacific Deep Slope Water<sup>51</sup> represents the main  
193 route for mid-depth flow in the South Pacific to enter the SO<sup>51,57,58</sup>. Volcanic activity at multiple sites  
194 along ridge systems (Fig. 1A), alongside complex interior circulation, results in widespread  $xs^3He$  in  
195 intermediate and deep waters ( $\sim 1000-4000$  m) in the South Pacific Ocean<sup>46</sup>. A previous meridional  
196 transect along  $88^\circ W$  shows the southward extension of  $xs^3He$  towards our study area (Fig. S3A), which  
197 could indicate plumes originating from ridges to the north e.g., the Sala Y Gomez Ridge and Chile Rise  
198 (Fig. 1A). Similarly, a zonal transect along  $54^\circ S$  shows an eastward extension of  $xs^3He$  from the South  
199 EPR (SEPR) that intersects the OOI station (Fig. 3D; Fig. S3B). Because the  $xs^3He$  observations do not  
200 allow us to discern a clear source region, we undertook trajectory tracking simulations to provide an  
201 independent estimate for the origin of the hydrothermal signal.

202 To trace trajectories from potential sources, a series of model release sites were first selected.  
203 Along the poorly-explored southern ridge systems, bathymetry along with  $xs^3He$  observations and  
204 previous literature<sup>33</sup> were used to select an arc of potential release sites to the north and west of station  
205 OOI. To simulate a neutrally buoyant hydrothermal plume, trajectories were initiated within a  $0.5^\circ$  grid

206 around an assumed source extending 500 m upwards in the water column above the ridge at the West  
207 Chile Rise (WCR), East Chile Rise (ECR), North EPR (NEPR), South EPR (SEPR) and PAR South (PARS)  
208 locations (Fig.1; Table 1) and tracked forwards in time. A reverse simulation was also performed  
209 whereby trajectories were backtracked from within a 0.5° grid around the OOI site between 2000-3950  
210 m depth, coincident with our hydrothermal Fe and Mn enrichment observations (Table 1; Fig.2C),  
211 towards the ridge systems. The mid-ocean ridge system surrounding our study region was divided into 5  
212 regions (Fig. S4, Table S4), including an additional region PAR North (PARN) which did not have a  
213 forward-tracking release site. We quantified the backwards trajectories that passed through the likely  
214 plume depths (i.e. extending up to 500 m of water column above a ridge<sup>59-61</sup>) in these regions (Table 1  
215 and S2). For both forward and backwards tracking, trajectories were traced for 20 years by applying a  
216 Lagrangian simulator, called 'parcels' (<https://oceanparcels.org/><sup>62</sup>), to the velocity components of a  
217 global ocean model (Nucleus for European Modelling of the Ocean; NEMO)<sup>63</sup> at an eddy-resolving  
218 (0.083°) horizontal resolution. Trajectories estimate the flow of water within the model ocean and do  
219 not represent concentrations of trace metals.

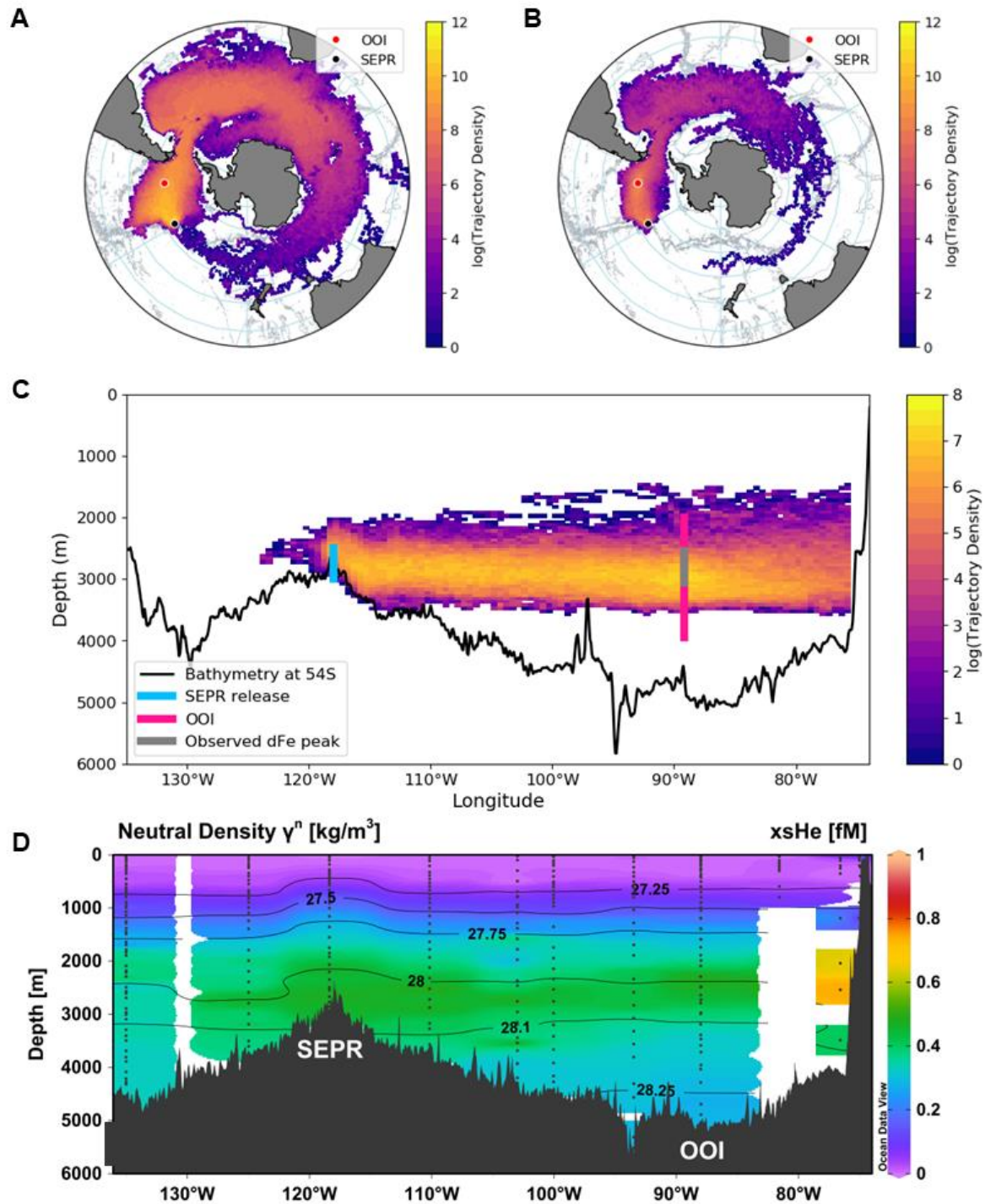
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221

222 **Table 1.** Summary of trajectory simulations used to identify possible hydrothermal source regions. The proportion of trajectories that passed  
 223 within a 0.5° area around the OOI site are presented relative to the number of particles that were successfully released during each simulation  
 224 (Table S2 and S3). The source regions that trajectories were tracked backwards in time towards were much larger than the area around station  
 225 OOI, as they span along sections of the ridges, allowing more trajectories to pass into the source regions in the backward-tracking simulations  
 226 (Table S2, S3 and S4).

Experiment type	Trajectory starting point	Observed signal	Ridge depth (m)	Trajectory seeding depth range (m)	Proportion of trajectories that passed near observed signal (%)	Median first passage time (years from start)
Forward	ECR	OOI	3900	3400-3900	0	>20
Forward	WCR	OOI	3900	3400-3900	0	>20
Forward	SEPR	OOI	3000	2500-3000	9.5	9.8
Forward	NEPR	OOI	3200	2700-3200	0.3	17.9
Forward	PARS	OOI	2800	2300-2800	0.9	15.1
Experiment type	Trajectory starting point	Source region	Ridge depth (m)	Plume source depth range (m)	Proportion of trajectories that passed near potential source (%)	Median first passage time (years from start)
Backward	OOI	ECR/WCR	3900	3400-3900	0	>20
Backward	OOI	SEPR	3000	2500-3000	20.7	11.8
Backward	OOI	NEPR	3200	2700-3200	1.3	14.4
Backward	OOI	PARN	3000	2500-3000	20.3	12.7
Backward	OOI	PARS	2800	2300-2800	7.7	14.4

227



229

230 **Figure 3.** Trajectories of the forward-tracking simulations from the SEPR (A, B, C) and coincident  $\text{xs}^3\text{He}$ 

231 anomaly (D). Trajectories were released between the ridge and 500 m above to mimic a neutrally-

232 buoyant hydrothermal plume. **A** Trajectory density, which indicates the number of instances of  
233 trajectories passing into a grid cell (log scale), of all SEPR trajectories. **B** Trajectory density map of SEPR  
234 trajectories that pass within a 0.5° area around the OOI station (red marker). **C** Depth section of  
235 trajectory density for the SEPR trajectories that pass within a 0.5° area of the OOI station (location  
236 indicated by pink line). Additionally, the grey line superimposed on the pink line highlights the depth  
237 range where dFe concentrations were >0.75 nM (Fig. 2). The cyan line marks the trajectory seeding  
238 depth which mimics a neutrally-buoyant plume. Bathymetry along 54°S (to match station OOI) is shown.  
239 Note the colorbar scale is different for **C** compared to **A** and **B**. **D** An east-west transect of excess Helium  
240 ( $\text{x}\text{s}^3\text{He}$ ) at 54°S as an indicator of hydrothermal influence from the Jenkins et al. (2019) dataset<sup>52</sup>.

241 Trajectory estimates strongly supported a westerly source region with no influence from  
242 northerly ridge systems (Table 1). The SEPR region is identified as the most likely source for our  
243 observed signal. Forward-tracking simulations from the SEPR resulted in the greatest percentage of  
244 trajectories (9%) crossing within a 0.5° area around OOI at a depth coincident with the observed Fe and  
245 Mn enrichment compared to other release sites (Figure 3C; blue line). Similarly, in the backward-  
246 tracking simulations, the greatest percentage of trajectories (21%) released from OOI passed within the  
247 SEPR source region (Table 1). In an oceanographic context, these percentages can be thought of as  
248 demonstrating the strength and coherency of transport pathways within the wider ocean circulation  
249 between the source regions and OOI i.e. the greater the percentage the more dominant the pathway.  
250 The SEPR also had the shortest median transit timescales to/from OOI (10-12 years). Further west, the  
251 backward-tracking simulations indicate that 8-20% of trajectories starting at OOI intersect with the PAR  
252 ridge systems with a median transit time of 13-14 years. However, the peak in the trajectory signal  
253 propagated from the PAR regions were deeper than the observed Fe and Mn peaks at OOI, especially for  
254 PARS (Figure S3H). Our transit time estimates from the SEPR and PAR are at the lower end of estimated  
255 hydrothermal dFe residence times of 9-50 years<sup>39,42</sup>.



256           The NEPR makes a minor contribution in both forward and backward trajectories over 20 years  
257 (0.3-1.3% of trajectories to/from OOI), with trajectories exhibiting longer median transit times to reach  
258 OOI (14-18 years), despite being much closer in proximity to OOI than the PARS site which made the  
259 second largest contribution (Fig. 1; Table 1). Similarly, trajectories from the Chile Rise did not intersect  
260 with the OOI site within 20 years.

261           Our simulations indicate minimal exchange between waters north and south of 45°S in the MAP,  
262 likely due to the South Pacific Current, which has diverging pathways above and below 40-43°S<sup>57,64</sup>. This  
263 interpretation is supported by a model validation between the NEMO model used for the trajectory  
264 simulations and the Estimating the Circulation and Climate of the Ocean (ECCO) model state  
265 estimate<sup>65,66</sup> (Figure S5). Both showed that the dominant currents at OOI originate from the west with  
266 weaker currents north of 45°S. Overall, the trajectory simulations indicated that the observed OOI trace  
267 metal plume originates from a ridge to the west of OOI, which is consistent with our understanding of  
268 the fast-moving ACC and weak currents in the northern MAP<sup>67</sup>. Therefore, the strong  $x\text{s}^3\text{He}$  signal to the  
269 north in the MAP region (Fig. S3A) should not be interpreted as traversing southwards towards OOI. Our  
270 results indicate that the  $x\text{s}^3\text{He}$  signal observed at OOI has most likely traversed eastwards from the SEPR  
271 and/or from along the PAR (Fig. 3D). The PARS/N trajectory simulations highlight a pathway for potential  
272 hydrothermal transport along the PAR, that tracks northwards up to the SEPR and then eastwards  
273 towards the OOI site (Fig. S4E, G). Thus, it is possible we observed the amalgamation of trace metal  
274 inputs from multiple vent sites along the PAR and SEPR at the OOI site and along the transect, rather  
275 than a discrete single vent signal.

276           Systematic surveying for hydrothermal activity is lacking for the majority of the southern EPR  
277 and PAR systems<sup>68</sup>. However, it is expected that the ridge system is hydrothermally active as multiple  
278 lines of evidence suggest that hydrothermal venting is common. Metalliferous sediments (enriched in Fe  
279 and Mn compared to aluminium), formed by precipitation of Fe and Mn from hydrothermal fluids, are

280 found along these ridge systems<sup>69</sup>. Additionally, the magmatic budget hypothesis<sup>68</sup> predicts that  
281 variability in magma supply is the primary control on the large-scale hydrothermal distribution pattern  
282 along spreading ridges and is supported by a linear relationship between spreading rate and frequency  
283 of vent fields. Extrapolation of this relationship has been used to estimate a total of ≈200-300  
284 undiscovered vent fields along the SEPR and PAR with an average distance between vent fields along the  
285 SEPR of 25-30 km<sup>68</sup>. Indeed, high resolution optical and redox sensor measurements made along 1470  
286 km of intermediate and fast spreading mid-ocean ridge suggest that the frequency of vent sites is 3 to 6-  
287 fold higher than current observations (Fig. 1A), with a mean discharge spacing of 3-20 km<sup>70</sup>.

288 Similarly, evidence from observational and modelling studies exists for the long-range transport  
289 of chemical signatures from these ridge systems. For example, sections of the PAR (south of 55°S) were  
290 identified as the likely source region of a <sup>3</sup>He anomaly found on the neutral density surface  $\Upsilon_n = 28.2 \text{ kg}$   
291  $\text{m}^{-3}$ , indicative of active venting<sup>33</sup>. Modelling dFe hydrothermal vent input as a function of ridge  
292 spreading rate found good agreement with observed dFe anomalies in the abyssal SO, with some of the  
293 highest rates of hydrothermal Fe input in the Pacific sector along the PAR and SEPR<sup>39</sup>. Similarly, a global  
294 ocean modelling simulation of Mn predicted strong input from the SEPR which produced a dMn anomaly  
295 that extends to the location of our transect<sup>12</sup>.

296

### 297 **2.3 Could hydrothermal iron and manganese from southeast Pacific vents fuel Southern** 298 **Ocean primary production?**

299 The potential for hydrothermal Fe and Mn to fuel primary production depends on the balance  
300 between Fe and Mn residence times and the ventilation timescale of hydrothermal plume-influenced  
301 waters. A transit time of  $99 \pm 18$  years for the shoaling of deep waters to the SO surface has been  
302 estimated using a global <sup>3</sup>He mass balance model, in which <sup>3</sup>He predominantly enters into the SO via  
303 deep South Eastern Pacific waters<sup>46</sup>. Deep ocean re-exposure timescales of a few centuries have also

304 been calculated using a low horizontal resolution ( $2^\circ$ ) global steady-state ocean circulation inverse  
305 model<sup>43</sup>. These ventilation timescales are likely incompatible with the notion that Fe and Mn from  
306 southeast Pacific vents could sustain significant amounts of Southern Ocean productivity when  
307 compared with estimated seawater residence times for Fe and Mn. Indeed, coupling a mechanistic Fe  
308 model, where hydrothermal input is estimated from a fixed Fe:<sup>3</sup>He ratio, with an ocean inverse  
309 circulation model allows for scavenging processes to effectively trap hydrothermal Fe in the deep  
310 ocean<sup>40</sup>.

311           However, uncertainty around the magnitude of hydrothermal Fe input from SO ridge systems,  
312 and accounting for mesoscale processes when estimating ventilation timescales, may in fact allow for  
313 rapid transport of hydrothermal Fe and Mn to upper ocean waters. An inverse modelling approach  
314 reduces the magnitude of <sup>3</sup>He input from SO ridge systems relative to a spreading rate model approach.  
315 A spreading rate model estimates <sup>3</sup>He input as a function of ridge spreading rate, and likewise assigns an  
316 Fe:<sup>3</sup>He ratio to estimate a hydrothermal Fe flux<sup>39</sup>. Significantly, the inverse model approach does not  
317 replicate the magnitude of an observed SO hydrothermal from the GEOTRACES GS01 section South of  
318 Tasmania<sup>39</sup>. A consequence of the spatial redistribution of hydrothermal Fe input away from the SO vent  
319 sites by the inverse modelling approach is a reduction in the estimated amount of hydrothermal Fe  
320 reaching the upper 250 m of the ocean by 4-5-fold relative to the spreading rate model<sup>40</sup>. It is also  
321 possible that previous studies may be over-estimating ventilation times due to their coarse resolution.  
322 Mesoscale (10-100 km) processes are important conduits for vertical mixing in the SO<sup>44,45</sup>. Using an  
323 eddy-resolving Lagrangian particle tracking model transit times of 17-90 years were estimated for CDW  
324 (originating from 30°S between 1000-4000 m) to upwell to the SO mixed layer<sup>44,45</sup>. Importantly, transit  
325 time estimates decrease as model resolution becomes finer<sup>44,45</sup> with the shorter 17 year timescale  
326 estimated using a 0.1 degree eddy-resolving model. This highlights that accounting for mesoscale eddies  
327 is important for estimates of timescales for SO upwelling<sup>45</sup>.

328 To investigate the potential fate of the observed dFe and dMn signals, we continued to forward-  
329 track trajectories that passed through our cruise transect at depths of the observed trace metal peak  
330 (2000-4000 m; Fig. 4). We thus estimated the proportion of trajectories which shoal to depths  
331 coincident with mixed layer depths associated with SO winter mixing, the key seasonal supply  
332 mechanism of micronutrients to SO surface waters<sup>35</sup>. 83% of the trajectories passing through the cruise  
333 transect between 2000-4000m continued east through Drake Passage and then flowed northward away  
334 from the polar front ( $\sim 60^\circ\text{S}$ ) into deep water masses (Fig. 4A; median depth 2087 m), and so are  
335 unlikely to supply hydrothermal Fe and Mn to the SO mixed layer. However, trajectories which pass  
336 through Drake Passage and traverse pathways south of  $60^\circ\text{S}$  (17%) tended to shoal in the water column  
337 by the end of the 20-year simulation (median depth 1043 m). Of the 17% of trajectories that passed  
338 south of  $60^\circ\text{S}$ , 10% (i.e., 1.7% of total trajectories) reached depths shallower than 600 m, a proxy for the  
339 winter maximum mixed layer depth in deep mixing regions of the SO<sup>71</sup>, while 31% (i.e. 5.3% of total)  
340 reached shallower than 1000 m, the maximum observed winter mixed layer depth<sup>71</sup>.

341 Our simulated trajectories thus indicate pathways from the observed deep southeast Pacific  
342 sector of the SO to the SO mixed layer within 20 years. Furthermore, the number of trajectories south of  
343  $60^\circ\text{S}$  reaching winter mixing depths are likely to continue to increase beyond the temporal limit of our  
344 simulations (20 years). As discussed above, resolving mesoscale features in high-resolution models, as in  
345 our study ( $0.083^\circ$  horizontal resolution), results in SO ventilation timescale estimates (within 20 years)  
346 that allow for hydrothermal Fe and Mn from SO vents to reach SO mixed layer depths within Fe and Mn  
347 residences times (5-50 years)<sup>42</sup>. Moreover, the effects of submesoscale mixing and dispersion, which will  
348 influence the transport and shoaling rates of dissolved constituents, are not resolved in the physical  
349 model driving our trajectory calculations but could further shorten ventilation timescales<sup>72</sup>. Trajectory  
350 modelling offers an alternate approach to identifying hydrothermal source regions and ventilation  
351 pathways and timescales, which may be more tractable for localised studies. It remains challenging to

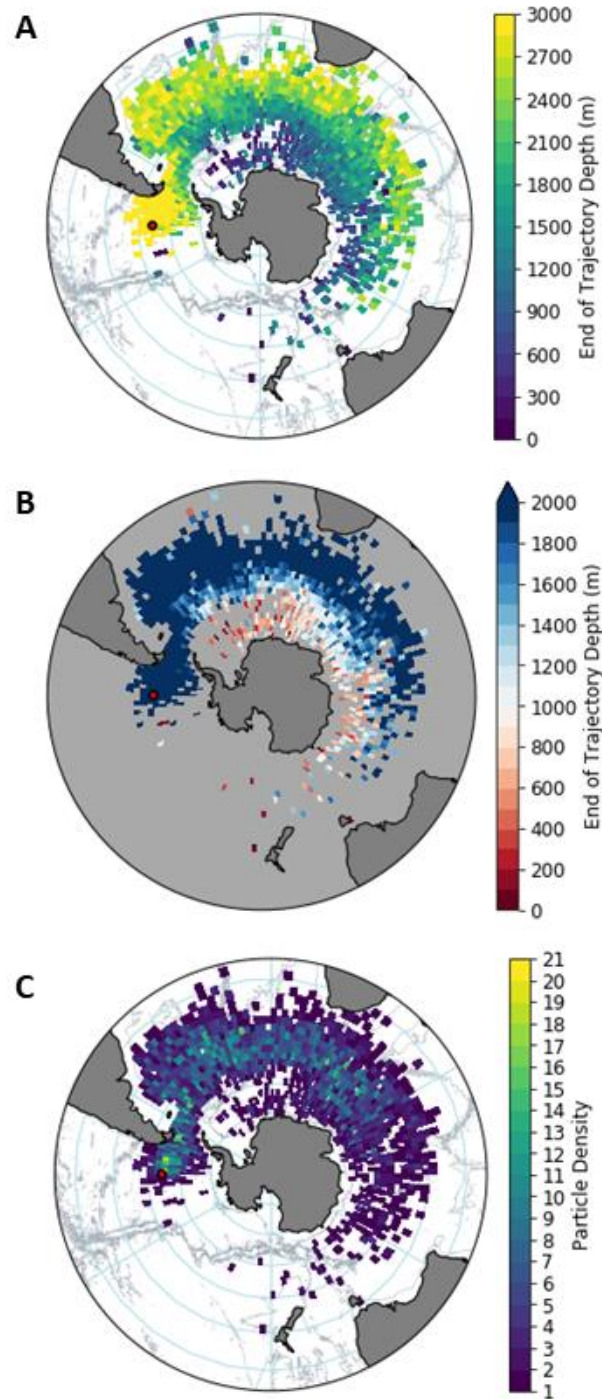
352 isolate what drives the differences between Fe supply from different modelling approaches<sup>39,40</sup> and  
353 ventilation timescales from high-resolution studies (<20 years)<sup>44,45</sup>, compared to coarse-resolution or  
354 global inventory studies (~100 years)<sup>43,46</sup> discussed above, as several factors change in tandem. The  
355 significance of transport pathways to the mixed layer must ultimately depend on the residence times of  
356 hydrothermal Fe and Mn being comparable or longer than the ventilation timescales we predict, as well  
357 as the consistency and stability of hydrothermal Fe and Mn inputs, both of which remain poorly  
358 constrained.

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364 **Figure 4.** Indication of trajectory locations at the end of the 20 years simulation from the source regions.

365 **A** Trajectory depth and location at 20 years and **B** the depths presented as above (red) and below (blue)

366 the maximum winter MLD in the SO (1000m) and **C** Trajectory density at 20 years. OOI is shown by the  
367 red marker. Note that only trajectories that passed within the cruise transect (OOI to 60°S) and the  
368 observed hydrothermal signal depth range (2000-4000 m) are shown.

### 369 **3.0 Conclusion**

370 We identify potential pathways for Fe and Mn from SO hydrothermal vents to reach the SO mixed layer.  
371 Enhanced Fe and Mn concentrations were observed between 2000-4000 m depths within the southeast  
372 Pacific sector of the SO. The observed trace metal enrichment most likely originated from the South East  
373 Pacific Rise (south of 30°S). An origin further westward, such as from the Pacific Antarctic Ridge, was  
374 also possible. We further identified pathways for rapid (< 20 years) transport of water parcels enriched  
375 in hydrothermal Fe and Mn, from vents located within the understudied Pacific sector of the SO, to SO  
376 winter mixed layer depths. Consensus on the role of deep ocean hydrothermal trace metal inputs in  
377 fuelling upper ocean primary production has not yet been reached, with modelling studies disagreeing  
378 on the importance of SO vent systems for supplying hydrothermal Fe supply to the wider SO euphotic  
379 zone<sup>39,40</sup>. However, we conclude that due to the vigorous action of the ACC, Pacific sector SO  
380 hydrothermal inputs provide the potential for sustained input of Fe and Mn to the euphotic zone south  
381 of the Antarctic Polar Front and far downstream of the deep ocean vent sites, potentially fuelling  
382 productivity in remote areas of the SO. Given the existence of ridge systems throughout the SO, it is  
383 likely that other similar pathways exist that rapidly transport Fe and Mn from vent sites to the SO  
384 euphotic zone.  
385



## 386 **Methods**

### 387 *Trace Metal and Macronutrient Methods*

388 Sampling was conducted during December 2019 and January 2020 on board the *R.R.S. Discovery* along a  
389 transect in the eastern Pacific sector of the SO (Fig.1). All trace metal samples were collected following  
390 GEOTRACES protocols<sup>73</sup>.

391 Briefly, dissolved Fe (0.2  $\mu\text{m}$  filtered) and TdFe (unfiltered) were analyzed using flow injection with  
392 chemiluminescence detection, after spiking with hydrogen peroxide<sup>74</sup>, in a clean laboratory at the  
393 University of Plymouth. The mean limit of detection was  $0.012 \pm 0.009$  nM and limit of quantification  
394  $0.038 \pm 0.026$  nM (n=54). Additional seawater samples collected in  $>1$  L volumes, and acidified for  $> 6$   
395 months, were used as in-house quality control materials as a measure of precision and were analysed  
396 every 10 samples. 4 in-house quality control materials were analysed; #1  $0.17 \pm 0.01$  nM (n=45), #2  $0.22$   
397  $\pm 0.04$  nM (n=54), #3  $0.26 \pm 0.03$  nM (n=124) and #4  $0.19 \pm 0.03$  nM (n=70). GEOTRACES GSP and SAFe  
398 D1 and S consensus values were analysed to determine accuracy, consensus values are (GSP  $0.16 \pm 0.05$   
399 nM, D1  $0.65 \pm 0.04$  nM, S  $0.10 \pm 0.01$  nM), which compared well to values determined (GSP  $0.20 \pm 0.020$   
400 nM, D1  $0.66 \pm 0.066$ , S  $0.10 \pm 0.01$  nM). The authors have previously published bottom up and top-down  
401 combined analytical uncertainty estimates ( $u_c$  5-10 % (k=1)) for this technique<sup>75,76</sup>.

402 A subset of samples from the northern, middle, and southern stations were chosen for further trace  
403 metal analysis. Dissolved Mn (0.2  $\mu\text{m}$  filtered) concentration was determined using a standard addition  
404 method with off-line pre-concentration and subsequent high resolution ICP-MS<sup>29</sup> at the National  
405 Oceanography Centre, Southampton, UK. The mean limit of detection was  $0.011 \pm 0.004$  nM (n=3).  
406 Consensus values for SAFe S ( $0.812 \pm 0.062$  nM) and D2 ( $0.360 \pm 0.051$  nM) reference material compared  
407 well with our measured values ( $0.769 \pm 0.059$  nM and  $0.372 \pm 0.030$  nM, respectively).

408 *Inorganic Nutrient Analysis*

409 Seawater samples were analysed for nitrate (determined as nitrate + nitrite;  $\text{NO}_3^- + \text{NO}_2^-$ ), silicate ( $\text{SiO}_4^{4-}$ ),  
410 nitrite ( $\text{NO}_2^-$ ) and phosphate ( $\text{PO}_4^{3-}$ ). Samples were drawn from Niskin bottles into rinsed 15 mL  
411 centrifuge tubes. Colorimetric analysis was undertaken within 24 hours of the samples being taken and  
412 performed using SEAL QuAAtro 39 segmented flow autoanalyser<sup>77</sup>. Two sets of certified reference  
413 materials (CRM Lots CJ and CB, KANSO, Japan) were determined at a start and the end of each sample  
414 run to ensure accuracy. Depending on an analysis run, the detection limits for each parameter ranged as  
415 follows: 0.02-0.2  $\mu\text{M}$  for nitrate + nitrite, 0.02-0.15  $\mu\text{M}$  for silicate, 0.01-0.033  $\mu\text{M}$  for nitrite, and 0.002-  
416 0.019  $\mu\text{M}$  for phosphate.

417

418 *Temperature and Salinity Calibration*

419 Trace-metal clean samples were taken with a titanium frame Conductivity Temperature Depth (CTD) fitted  
420 with 10 L Niskin bottles and a SBE 9 plus underway unit. Conductivity (and calculated salinity) data were  
421 calibrated against samples taken from each cast and run on an Autosal 8400B salinometer. The salinity  
422 was accurate to within  $\pm 0.004$ . There was no SBE35 thermometer fitted on the titanium frame but  
423 comparison with co-located calibrated stainless steel casts showed good agreement and data is accurate  
424 to better than  $\pm 0.005^\circ\text{C}$

425

426 *Excess Helium Calculations*

427  $^3\text{He}$  is a primordial substance trapped in the mantle during planetary formation. Ratios of  $^3\text{He}/^4\text{He}$  in  
428 excess of atmospheric values indicate a contribution from the mantle, therefore  $^3\text{He}$  can be used to  
429 identify ocean waters that have been influenced by hydrothermal activity. Excess  $^3\text{He}$  is the approximate

430 measure of non-atmospheric  $^3\text{He}$  over saturation, we followed the approach outlined in Resing et al.  
431 (2015)<sup>23</sup>:

$$432 \quad {}^3\text{He}_{\text{xs}} = (\delta^3\text{He} - \delta^{3*}\text{He}) / 100 \times C[\text{He}] \times 1.384 \times 10^{-6}$$

433 where  $\delta^3\text{He} = 100 \times (\text{Rx}/\text{Ra} - 1) \times 100\%$ , Rx and Ra are the  $^3\text{He}/^4\text{He}$  ratios of the sample and air ( $1.384 \times$   
434  $10^{-6}$ ), respectively.  $\delta^{3*}\text{He}$  is the helium isotope ratio anomaly in solubility equilibrium with the  
435 atmosphere.

436

### 437 *Trajectory Modelling*

438 To identify the potential source of the observed hydrothermal Fe and Mn signal we focus on regions  
439 along the Chile Rise (CR), East Pacific Rise (NEPR and SEPR) and Pacific-Antarctic Ridge (PARN and PARS),  
440 with regions of suspected hydrothermal activity shown in Fig. 1A (triangles).

441 Trajectories were used to identify the region(s) that were most likely to be the source of the deep water  
442 hydrothermal iron signal. Trajectories were tracked forwards from possible vent sites which may act as a  
443 source, and backwards from the OOI sustained observatory site, where enrichments in midwater  
444 micronutrient concentrations were observed. Trajectories do not represent quantities or concentrations  
445 of dFe or dMn. The velocity components used to calculate trajectories do not represent submesoscale  
446 mixing or dispersion processes<sup>62</sup>, which may affect the pathway of dissolved constituents in the ocean.

447 We estimated 20-year trajectories using ‘parcels’ (version 2.2.0; <https://oceanparcels.org/><sup>62</sup>). Parcels  
448 was applied to the NEMO eddy-resolving general circulation model (ORCA0083-N06<sup>63</sup>). Model output at  
449  $1/12^\circ$  horizontal resolution from 1992-2011 was used as a 20-year climatology of the hydrodynamic flow  
450 field. NEMO was coupled to a sea-ice model (LIM2<sup>78</sup>) and was forced with historical atmospheric  
451 reanalysis (Drakkar Forcing Set 5.2<sup>79</sup>). The simulations were carried out using 3D Runge-Kutta fourth

452 order (RK4) timestep integration as the advection scheme with a timestep of 1 day to estimate the  
453 trajectories<sup>80</sup>. The trajectory position was exported every 5 days.

454 Applying 'parcels' to velocity components on a C grid whilst using an RK4 advection scheme, with a  
455 Lagrangian timestep of 1 day, can lead to trajectories 'overshooting' the ocean grid cells into  
456 bathymetry. To reduce the occurrences of particles travelling out of bounds we added a criterion that  
457 returned such trajectories to 10 m above the seafloor<sup>81</sup>. This criterion was applied to 4.7% of the total  
458 timesteps in the backwards-tracked trajectories and for an average of 22.9% of timesteps for the  
459 forward-tracked trajectories, due to the trajectory start locations being in close proximity to seafloor  
460 topography, i.e. vent sites along ocean ridges.

#### 461 *Forward-tracking Trajectories*

462 Six potential source regions were identified (Fig. 1A, triangles) and referred to as the East Chile Rise  
463 (ECR), West Chile Rise (WCR), Easter Microplate (EM), North East Pacific Rise (NEPR), South East Pacific  
464 Rise (SEPR) and the Pacific Antarctic Ridge (PAR; Table S2). Trajectories were initialised over a 0.5° grid  
465 around each potential source location at 100 m intervals between 2000-4000 m. 2000 trajectories were  
466 initiated per depth. In regions of bathymetry shallower than 4000 m, fewer trajectories were tracked. All  
467 calculations were normalised to the number of successfully tracked trajectories (Table S2) that were  
468 within a plume. We define the plume by the deepest depth of successful trajectories initialisation within  
469 each 0.5° release area, to mimic the deepest possible vent depth, up to 500 m shallower in the water  
470 column. Trajectories that crossed within a 0.5° area around the OOI site were isolated to estimate the  
471 dilution percentage of trajectories from the source regions (trajectory start locations) to OOI (Table S3).  
472 The EM had no trajectories passing near the OOI site and was not included in the analysis (Table S2).

#### 473 *Backward-tracking Trajectories*

474 To complement the forward-tracking we initiated trajectories throughout the water column at OOI to  
475 coincide with the depths at which the dFe and dMn signal was observed. Trajectories were initialised  
476 over a 0.5° grid around the OOI sustained observatory site every 50m between 2000-4000 m. 1000  
477 trajectories were released per depth and tracked backwards in time for 20 years (2011 to 1992; Table  
478 S2).

479 We defined 5 potential hydrothermal signal source regions along the Pacific Antarctic Ridge, East Pacific  
480 Rise and the Chile Rise to further evaluate where the iron signal was most likely to originate from – ECR  
481 and WCR as CR, NEPR, SEPR, Pacific Antarctic Ridge North (PARN), and Pacific Antarctic Ridge South  
482 (PARS; Table S4). Trajectories were only classed as originating from that source region if they entered at  
483 depths within a range of 500 m above the deepest possible vent height within each release area to  
484 mimic a plume<sup>82,83</sup>. For PARN, which had no specific vent release site for the forward tracking simulation,  
485 a vent depth of 2500 m was chosen by examining the depths of the topography within the PARN area as  
486 a histogram to identify the ridge depths. The fraction of trajectories passing into each source region is  
487 presented as the dilution percentage (Table 1).

488

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498

499 **Data Statement**

500 Lagrangian trajectories from the tracking using parcels are available at Zenodo  
501 (DOI:10.5281/zenodo.8153763). The NEMO model velocity components used to force parcels can be  
502 found on JASMIN (<https://www.ceda.ac.uk/services/jasmin/>). The micronutrient data has been  
503 submitted to the British Oceanographic Data Centre (DOI: 10.5285/0c7760cc-98fb-5861-e063-  
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